

Part One

Background

1

Petroleum Refining and Petrochemical Industry Overview

Petroleum refining and the petrochemical industry account for a major share in the world energy and industrial market. In many situations, they represent the economic back-bone of industrial countries. Today, the volatile environment of the market and the continuous change in customer requirements lead to constant pressure to seek opportunities that properly align and coordinate the different components of the industry. In particular, petroleum refining and petrochemical industry coordination and integration is gaining a great deal of interest.

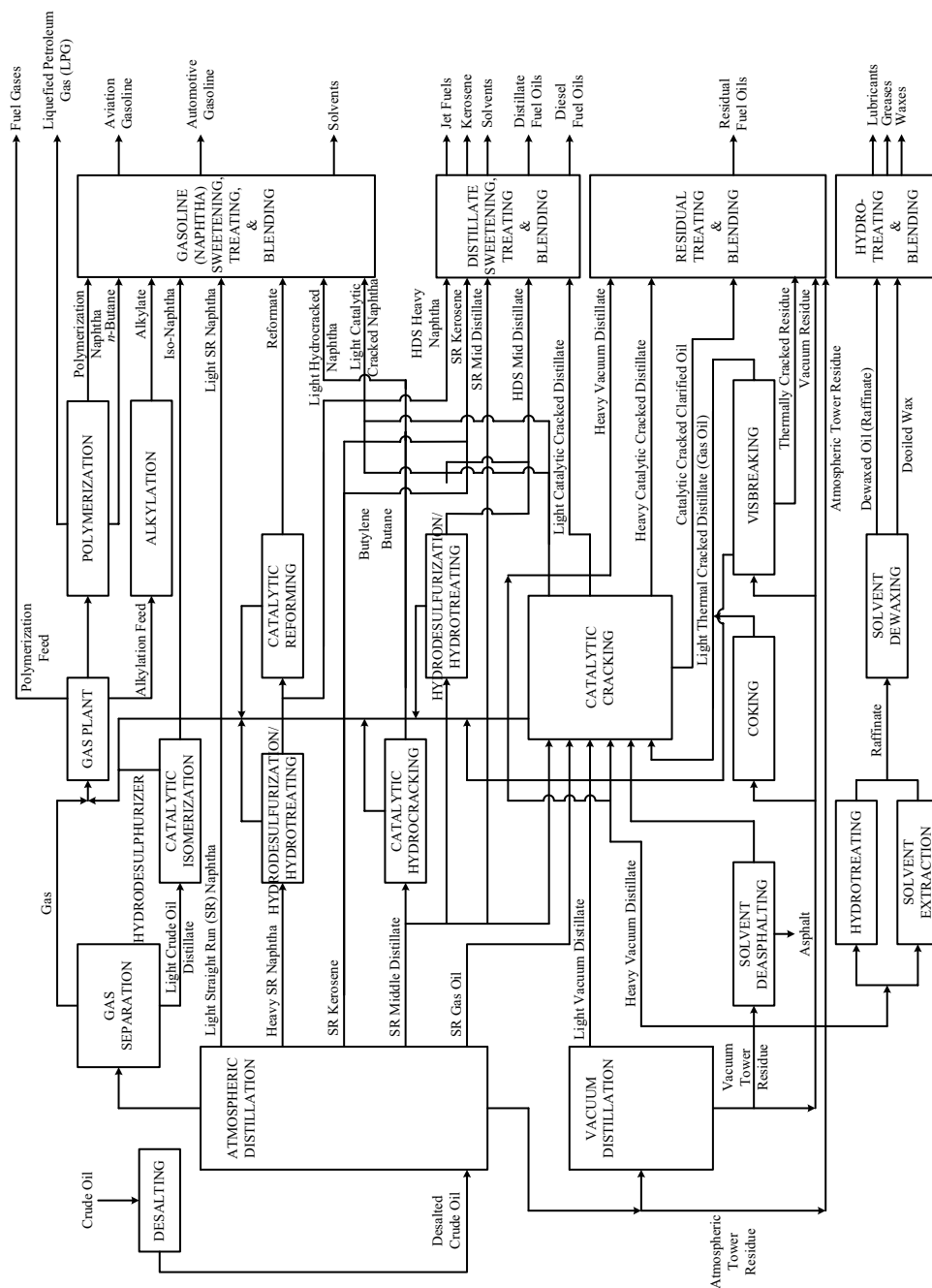
In this chapter, we will give an overview of the process configurations of petroleum refining and the petrochemical industry. We will also discuss the key classifications of petrochemical industry feedstock from petroleum products and explain and propose possible synergies between the petroleum refinery and the petrochemical industry.

1.1

Refinery Overview

The first refinery was built in Titusville, Pennsylvania in 1860 at a cost of \$15 000 (Nelson, 1958). This refinery and other refineries at that time only used batch distillation to separate kerosene and heating oil from other crude fractions. During the early years, refining separation was performed using batch processing. However, with the increase in demand for petroleum products, continuous refining became a necessity. The first widely recognized continuous refinery plants emerged around 1912 (Nelson, 1958). With the diversity and complexity of the demand for petroleum products, the refining industry has developed from a few simple processing units to very complex production systems. A simplified process flow diagram of a typical modern refinery is shown in Figure 1.1. For a detailed history of the evolution of refining technologies, we refer the reader to Nelson (1958) and Wilson (1997).

Typically, a refinery is made up of several distinct components that constitute a total production system, as shown in Figure 1.2. These components include:



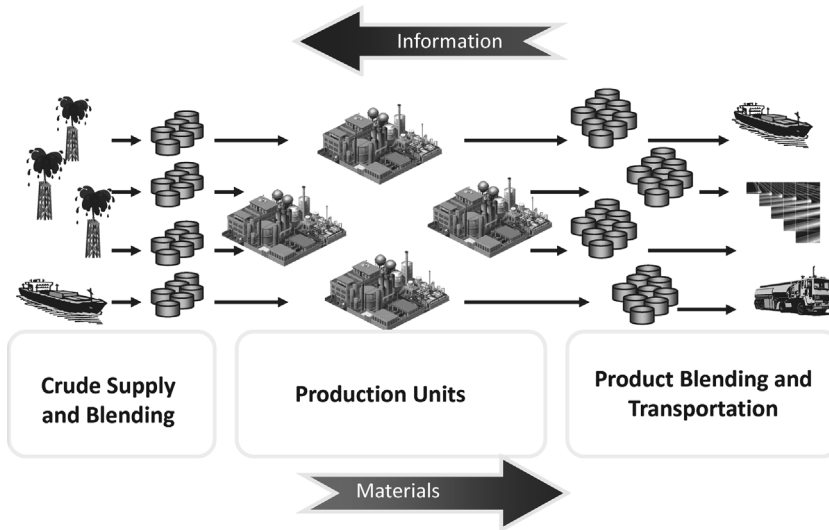


Figure 1.2 Schematic diagram of standard refinery configuration.

- **Crude Supply and Blending:** This area includes receiving facilities and a tank area (tank farm) where all crude oil types are received and either blended or sent directly to the production system.
- **Production Units:** Production units separate crude oil into different fractions or cuts, upgrade and purify some of these cuts, and convert heavy fractions to light, more useful fractions. This area also includes the utilities which provide the refinery with fuel, flaring capability, electricity, steam, cooling water, fire water, sweet water, compressed air, nitrogen, and so on, all of which are necessary for the safe operation of the refinery.
- **Product Blending and Transportation:** In this area the final products are processed according to either predetermined recipes and/or to certain product specifications. This area also includes the dispatch (terminals) of finished products to the different customers.

1.2

Mathematical Programming in Refining

The petroleum industry has long made use of mathematical programming and its different applications. The invention of both the simplex algorithm by Dantzig in 1947 and digital computers was the main driver for the widespread use of linear programming (LP) applications in the industry (Bodington and Baker, 1990). Since then, many early applications followed in the area of refinery planning (Symonds, 1955; Manne, 1958; Charnes and Cooper, 1961; Wagner, 1969; Addams and Griffin, 1972) and distribution planning (Zierer, Mitchell and White, 1976).

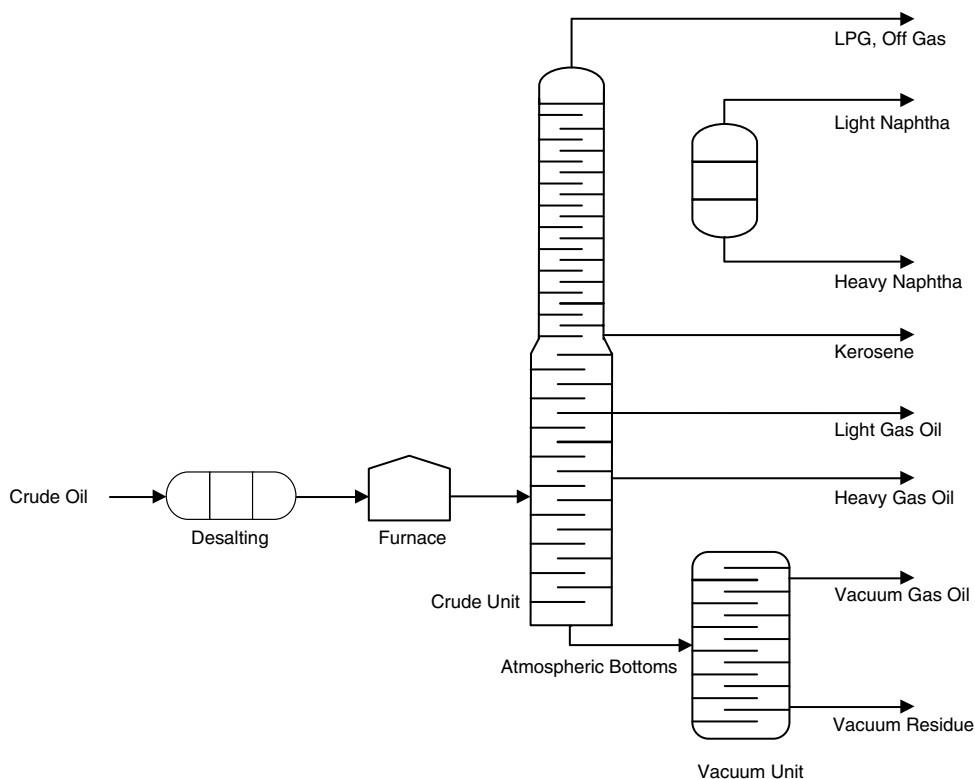


Figure 1.3 Process flow diagram of crude oil distillation process.

One of the main challenges that inspired more research in the area of refining was the blending or pooling problem (Bodington and Baker, 1990). The inaccurate and inconsistent results from the use of linear blending relations led to the development of many techniques to handle nonlinearities. The nonlinearities arise mainly because product properties, such as octane number and vapor pressure, assume a nonlinear relationship of quantities and properties of each blending component (Lasdon and Waren, 1983). In this context, we will describe two commonly used approaches in industry and commercial planning softwares to tackle this problem. They are linear blending indices and successive linear programming (SLP).

Linear blending indices are dimensionless numerical figures that were developed to represent true physical properties of mixtures on either a volume or weight average basis (Bodington and Baker, 1990). They can be used directly in the LP model and span the most important properties in petroleum products, including octane number, pour point, freezing point, viscosity, sulfur content, and vapor pressure. Many refineries and researchers use this approximation. Blending indices tables and graphs can often be found in petroleum refining books such as Gary and Handwerk (1994) or can be proprietorially developed by refining companies for their own use.

Successive linear programming, on the other hand, is a more sophisticated method to linearize blending nonlinearities in the pooling problem. The idea of SLP was first introduced by Griffith and Stewart (1961) of the Shell Oil Company where it was named the method of approximation programming (MAP). They utilized the idea of a Taylor series expansion to remove nonlinearities in the objective function and constraints then solving the resulting linear model repeatedly. Every LP solution is used as an initial solution point for the next model iteration until a satisfying criterion is reached. Bounding constraints were added to ensure the new model feasibility. Following their work, many improvement heuristics and solution algorithms were developed to accommodate bigger and more complex problems (Lasdon and Waren, 1980). Most commercial blending softwares and computational tools nowadays are based on SLP, such as RPMS by Honeywell Process Solutions (previously Booner and Moore, 1979) and PIMS by Aspen Technology (previously Bechtel Corp., 1993). However, such commercial tools are not built to support studies on capacity expansion alternatives, design of plants integration and stochastic modeling and analysis.

All in all, the petroleum industry has invested considerable effort in developing sophisticated mathematical programming models to help planners provide overall planning schemes for refinery operations, crude oil evaluation, and other related tasks.

1.3

Refinery Configuration

1.3.1

Distillation Processes

Crude oil distillation is the heart of and major unit in the refinery. Distillation is used to separate oil into fractions by distillation according to their boiling points. Prior to distillation, crude oil is first treated to remove salt content, if higher than 10 lb/1000 bbl, using single or multiple desalting units. This is required in order to minimize corrosion and fouling in the downstream heating trains and distillation columns. As illustrated in Figure 1.3, distillation is usually divided into two steps, atmospheric and vacuum fractionation according to the pressure at which fractionation is achieved. This is done in order to achieve higher separation efficiencies at a lower cost. After heating the crude to near its boiling point, it is introduced to the distillation column in which vapor rising through trays in the column is in direct contact with down-flowing liquid on the trays. During this process, higher boiling point fractions in the vapor phase are condensed and lighter fractions in the liquid are vaporized. This continuous process allows the various fractions of the crude oil with similar boiling points to achieve equilibrium and separate. Liquid can then be drawn off the column at different heights as product and sent for further treating or storage. Common products from the atmospheric distillation column include liquefied petroleum gas (LPG), naphtha, kerosene, gas oils and heavy residues.

The atmospheric bottom, also known as reduced oil, is then sent to the vacuum unit where it is further separated into vacuum gas oil and vacuum residues. Vacuum distillation improves the separation of gas oil distillates from the reduced oil at temperatures less than those at which thermal cracking would normally take place. The basic idea on which vacuum distillation operates is that, at low pressure, the boiling points of any material are reduced, allowing various hydrocarbon components in the reduced crude oil to vaporize or boil at a lower temperature. Vacuum distillation of the heavier product avoids thermal cracking and hence product loss and equipment fouling.

1.3.2

Coking and Thermal Processes

Nowadays more refineries are seeking lighter and higher quality products out of the heavy residues. Coking and other thermal processes convert heavy feedstocks, usually from distillation processes, to more desirable and valuable products that are suitable feeds for other refinery units. Such units include coking and visbreaking.

One of the widely used coking processes is delayed coking. It involves severe thermal cracking of heavy residues such as vacuum oil, thermal tars, and sand bitumen. The actual coking in this process takes place in the heater effluent surge drum and for this reason the process is called “delayed coking”. The coke produced by this process is usually a hard and porous sponge-like material. This type of coke is called sponge coke and exists in a range of sizes and shapes. Many other types of coke are commercially available in the market and have a wide range of uses, see Table 1.1. Other coking processes, including flexicoking and fluid coking, have been developed by Exxon.

The other thermal cracking process is visbreaking. This is a milder thermal process and is mainly used to reduce the viscosities and pour points of vacuum residues to

Table 1.1 End use of coke products (Gary and Handwerk, 2001).

Application	Coke type	End use
Carbon source	Needle	Electrodes
	Sponge	Synthetic graphite Aluminum anodes TiO ₂ pigments Carbon raiser Silicon carbide Foundries Coke ovens
Fuel use	Sponge	Space heating in Europe/Japan Industrial boilers
	Shot	Utilities
	Fluid	Cogeneration
	Flexicoke	Lime Cement

meet some types of fuel oil specifications and also to increase catalytic cracker feedstock. The two widely used processes in visbreaking are coil visbreaking and soaker visbreaking. In coil visbreaking most of the cracking takes place in the furnace coil whereas in soaker visbreaking, cracking takes place in a drum downstream of the heater, called the soaker. Each process offers different advantages depending on the given situation.

1.3.3

Catalytic Processes

There are two types of catalytic conversion units in the refinery, cracking and alteration processes. Catalytic cracking converts heavy oils into lighter products that can be blended to produce high value final products, such as gasoline, jet fuels and diesel. Whereas, catalytic altering processes convert feedstocks to higher quality streams by rearranging their structures. These processes include reforming, alkylation and isomerization units. Catalytic processes produce hydrocarbon molecules with double bonds and form the basis of the petrochemical industry.

1.3.3.1 Cracking Processes

Cracking processes mainly include catalytic cracking and hydrocracking. Catalytic cracking involves breaking down and rearranging complex hydrocarbons into lighter molecules in order to increase the quality and quantity of desirable products such as kerosene, gasoline, LPG, heating oil, and petrochemical feedstock. Catalytic cracking follows a similar concept to thermal cracking except that catalysts are used to promote and control the conversion of the heavier molecules into lighter products under much less severe operating conditions. The most commonly used process in the industry is fluid catalytic cracking (FCC) in which oil is cracked in a fluidized catalyst bed where it is continuously circulated between the reaction state and the regeneration state.

Hydrocracking on the other hand is a process that combines catalytic cracking and hydrogenation where the feed is cracked in the presence of hydrogen to produce more desirable products. This process mainly depends on the feedstock characteristics and the relative rates of the two competing reactions, hydrogenation and cracking. In the case where the feedstock has more paraffinic content, hydrogen acts to prevent the formation of polycyclic aromatic compounds. Another important role of hydrogen is to reduce tar formation and prevent buildup of coke on the catalyst.

1.3.3.2 Alteration Processes

Alteration processes involve rearranging feed stream molecular structure in order to produce higher quality products. One of the main processes in this category is catalytic reforming. Reforming is an important process used to convert low-octane feedstock into high-octane gasoline blending components called reformate. The kinetics of reforming involves a wide range of reactions such as cracking, polymerization, dehydrogenation, and isomerization taking place simultaneously. Depending on the properties of the feedstock, measured by the paraffin, olefin, naphthene,

and aromatic content (PONA), and catalysts used, reformates can be produced with very high concentrations of toluene, benzene, xylene, and other aromatics. Hydrogen, a by-product of the reforming process, is separated from the products and used as a feed in other refining processes.

Another alteration process is alkylation which is used to produce higher octane aviation gasoline and petrochemical feedstock for explosives and synthetic rubber. An isomerization process is also used to produce more material as an alkylation feedstock.

1.3.4

Treatment Processes

Treatment processes are applied to remove impurities, and other constituents that affect the properties of finished products or reduce the efficiency of the conversion processes. A typical example of a treating process is hydrotreating.

Catalytic hydrotreating is a hydrogenation process used to remove about 90% of contaminants such as nitrogen, sulfur, oxygen, and metals from liquid petroleum fractions. These contaminants, if not removed from the petroleum fractions, can have a negative impact on the equipment, the catalysts, and the quality of the finished product. Hydrotreating is mainly used prior to catalytic reforming to reduce catalyst contamination and before catalytic cracking to reduce sulfur and improve product yields. It is also used to upgrade middle-distillate petroleum fractions into finished kerosene, diesel fuel, and heating fuel oils and converts olefins and aromatics to saturated compounds. One of the emerging applications of this process is the treatment of pyrolysis gasoline (pygas), a by-product from the manufacture of ethylene, in order to improve its quality. Typically, the outlet for pygas has been motor gasoline blending, a suitable route in view of its high octane number. However, only small portions can be blended untreated owing to the unacceptable odor, color, and gum-forming tendencies of this material. The quality of pygas, which is high in diolefin content, can be satisfactorily improved by hydrotreating, whereby conversion of diolefins into mono-olefins provides an acceptable product for motor gas blending.

1.3.5

Product Blending

Blending is the process of mixing hydrocarbon fractions, additives, and other components to produce finished products with specific properties and desired characteristics. Products can be blended in-line through a manifold system, or batch blended in tanks and vessels. In-line blending of gasoline, distillates, jet fuel, and kerosene is accomplished by injecting proportionate amounts of each component into the main stream where turbulence promotes thorough mixing. Additives, including octane enhancers, metal deactivators, anti-oxidants, anti-knock agents, gum and rust inhibitors, detergents, and so on, are added during and/or after blending to provide specific properties not inherent in hydrocarbons.

1.4

Petrochemical Industry Overview

The petrochemical industry is a network of highly integrated production processes. The products of one plant may have an end use but may also represent raw materials for another process. Most chemicals can be produced by many different sequences of reactions and production processes. This multiplicity of production schemes offers the opportunity for switching between production methods and raw materials utilization.

Petroleum feedstock, natural gas and tar represent the main production chain drivers for the petrochemical industry (Bell, 1990). From these, many important petrochemical intermediates are produced, including ethylene, propylene, butylenes, butadiene, benzene, toluene, and xylene. These essential intermediates are then converted to many other intermediates and final petrochemical products, constructing a complex petrochemical network. Figure 1.4 depicts a portion of the petrochemical alternative routes to produce cellulous acetate.

Figure 1.4 is in fact a small extract from much larger and comprehensive flow diagrams found in Stanford Research Institute (SRI) reports. Note that certain chemicals, acetaldehyde and acetic acid for example, appear in more than one place in the flowchart. This reflects the different alternative production routes available for most chemicals. In the industry, many chemicals are products of more than one

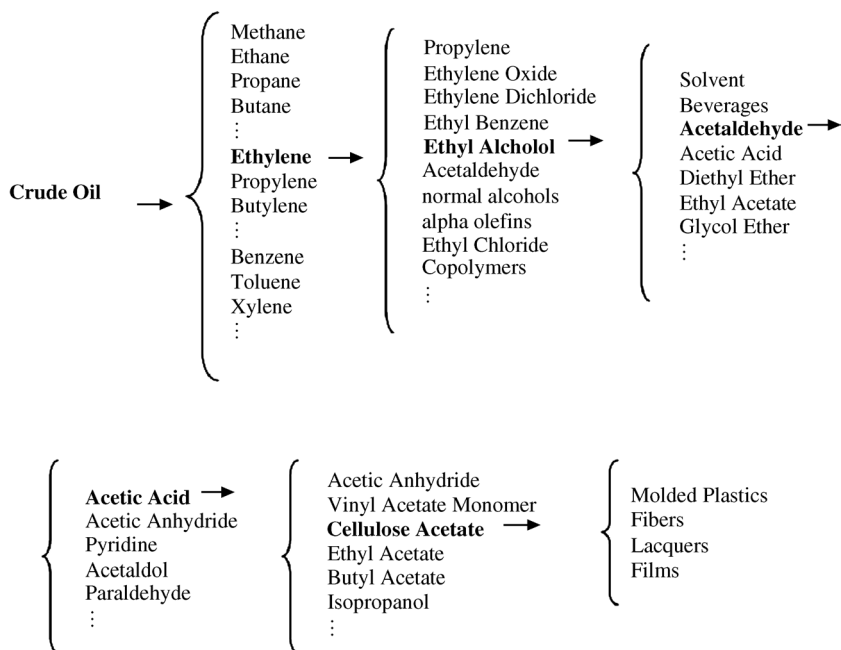


Figure 1.4 A Single Route of petroleum feedstock to petrochemical products (Bell, 1990).

process, depending upon local conditions, corporate policies, and desired by-products (Bell, 1990).

The flexibility in the petrochemical industry production and the availability of many process technologies require adequate strategic planning and a comprehensive analysis of all possible production alternatives. Therefore, a model is needed to provide the development plan of the petrochemical industry. The model should account for market demand variability, raw material and product price fluctuations, process yield inconsistencies, and adequate incorporation of robustness measures.

The realization of the need and importance of petrochemical planning has inspired a great deal of research in order to devise different models to account for the overall system optimization. Optimization models include continuous and mixed-integer programming under deterministic or parameter uncertainty considerations. Related literature is reviewed at a later stage in this book, based on the chapter topic.

1.5

Petrochemical Feedstock

The preparation of intermediate petrochemical streams requires different processing alternatives depending on the feedstock quality. In our classification of petrochemical feedstocks we closely follow that of Gary and Handwerk (1994) consisting of aromatics, olefins, and paraffin/cyclo-paraffin compounds. The classification of petrochemical feedstocks into these clusters helps to identify the different sources in the refinery that provide suitable feedstock and, therefore, to better recognize areas of synergy between the refinery and petrochemical systems.

1.5.1

Aromatics

Aromatics are hydrocarbons containing a benzene ring which is a stable and saturated compound. Aromatics used by the petrochemical industry are mainly benzene, toluene, xylene (BTX) as well as ethylbenzene and are produced by catalytic reforming where their yield increases with the increase in reforming severity (Gary and Handwerk, 1994). Extractive distillation by different solvents, depending on the chosen technology, is used to recover such compounds. BTX recovery consists of an extraction using solvents that enhances the relative volatilities of the preferred compound followed by a separation process based on the boiling points of the products. Further processing of xylenes using isomerization/separation processes is commonly required to produce *o*-, *m*-, and *p*-xylene mixtures, depending on market requirements. Benzene, in particular, is a source of a wide variety of chemical products. It is often converted to ethylbenzene, cumene, cyclohexane, and nitrobenzene, which in turn are further processed to other chemicals including styrene, phenol, and aniline (Rudd *et al.*, 1981). Toluene production, on the other hand, is mainly driven by benzene and mixed xylenes demand. Mixed xylene, particularly in Asia, is used to produce *para*-xylene and polyester (Balaraman, 2006).

The other source of aromatics is the pyrolysis gasoline (pygas) which is a byproduct of naphtha or gas oil steam cracking. This presents an excellent synergistic opportunity between refinery, BTX complex and steam cracking for olefins production.

1.5.2

Olefins

Olefins are hydrocarbon compounds with at least two carbon atoms and having a double bond. Their unstable nature and tendency to polymerize makes them one of the very important building blocks for the chemical and petrochemical industry (Gary and Handwerk, 1994). Although olefins are produced by fluid catalytic cracking in refineries, the main production source is through steam cracking of liquefied petroleum gas (LPG), naphtha or gas oils.

The selection of steam cracker feedstock is mainly driven by market demand as different feedstock qualities produce different olefins yields. One of the commonly used feed quality assessment methods in practice is the Bureau of Mines Correlation Index (BMCI) (Gonzalo *et al.*, 2004). This index is a function of average boiling point and specific gravity of a particular feedstock. The steam cracker feed quality improves with a decrease in the BMCI value. For instance, vacuum gas oil (VGO) has a high value of BMCI and, therefore, is not an attractive steam cracker feed. The commonly used feedstocks in industry are naphtha and gas oil.

Steam cracking plays an instrumental role in the petrochemical industry in terms of providing the main petrochemical intermediates for the downstream industry. The steam cracker olefin production includes ethylene, propylene, butylene and benzene. These intermediates are further processed into a wide range of polymers (plastics), solvents, fibers, detergents, ammonia and other synthetic organic compounds for general use in the chemical industry (Rudd *et al.*, 1981). In a situation where worldwide demand for these basic olefins is soaring, more studies are being conducted to maximize steam cracking efficiency (Ren, Patel and Blok, 2006). An alternative strategy would be to seek integration possibilities with the refinery as they both share feedstocks and products that can be utilized to maximize profit and processing efficiency.

1.5.3

Normal Paraffins and Cyclo-Paraffins

Paraffin hydrocarbon compounds contain only single bonded carbon atoms which give them higher stability. Normal paraffin compounds are abundantly present in petroleum fractions but are mostly recovered from light straight-run (LSR) naphtha and kerosene. However, the non-normal hydrocarbon components of LSR naphtha have a higher octane number and, therefore, are preferred for gasoline blending (Meyers, 1997). For this reason, new technologies have been developed to further separate LSR naphtha into higher octane products that can be used in the gasoline pool and normal paraffins that are used as steam cracker feedstock (e.g., UOP IsoSiv™ Process). The normal paraffins recovered from kerosene, on the other hand, are mostly used in biodegradable detergent manufacturing.

Cyclo-paraffins, also referred to as naphthenes, are mainly produced by dehydrogenation of their equivalent aromatic compounds; such as the production of cyclohexane by dehydrogenation of benzene. Cyclohexane is mostly used for the production of adipic acid and nylon manufacturing (Rudd *et al.*, 1981).

1.6

Refinery and Petrochemical Synergy Benefits

Process integration in the refining and petrochemical industry includes many intuitively recognized benefits of processing higher quality feedstocks, improving the value of byproducts, and achieving better efficiencies through sharing of resources. Table 1.2 illustrates different refinery streams that can be of superior quality when used in the petrochemical industry. The potential integration alternatives for refining and petrochemical industries can be classified into three main categories; (i) process integration, (ii) utilities integration, and (iii) fuel gas upgrade. The integration opportunities discussed below are for a general refinery and a petrochemical complex. Further details and analysis of the system requirements can be developed based on the actual system infrastructure, market demand, and product and energy prices.

1.6.1

Process Integration

The innovative design of different refinery processes while considering the downstream petrochemical industry is an illustration of the realization of refining and

Table 1.2 Petrochemical alternative use of refinery streams (Anon, 1998).

Refinery Stream	Petrochemical Stream	Alternative Refinery Use
FCC offgas	Ethylene	Fuel gas
Refinery propylene (FCC)	Propylene	Alkylation/polygasoline
Reformate	Benzene, toluene, xylenes	Gasoline blending
Naphtha and LPG	Ethylene	Gasoline Blending
Dilute ethylene (FCC and delayed coker offgas)	Ethylbenzene	Fuel gas
Refinery propylene (FCC product)	Polypropylene, Cumene, Isopropanol, Oligomers	Alkylation
Butylenes (FCC and delayed coker)	MEK (methyl ethyl ketone)	Alkylation, MTBE
Butylenes (FCC and delayed coker)	MTBE	Alkylation, MTBE
Refinery benzene and hydrogen	Cyclohexane	Gasoline blending
Reformate	o-xylene	Gasoline blending
Reformate	p-xylene	Gasoline blending
Kerosine	n-paraffins	Refinery product
FCC light cycle oil	Naphthalene	Diesel blending

petrochemical integration benefits. This is demonstrated by the wide varieties of refinery cracking and reforming technologies that maximize olefin production. Some of the available technologies include cracking for high propylene and gasoline production (Fujiyama *et al.*, 2005), maximum gasoline and LPG production, and low-pressure combination-bed catalytic reforming for aromatics (Wang, 2006). Other technologies include different extractive treatments of refinery streams, for example, aromatic recovery from light straight-run (LSR) naphtha. The normal paraffins of the LSR, on the other hand, are typically used as a steam cracker feedstock (Meyers, 1997).

Reforming, as mentioned above, is the main source of aromatics in petroleum refining where their yield increases with the increase in reforming severity. Aromatics in the reformat streams are recovered by extractive distillation using different solvents, depending on the chosen technology. The benzene-toluene-xylenes (BTX) complex is one of the petrochemical processes that makes use of many of the benefits of integration with petroleum refining. The integration benefits are not only limited to the process side but also extend to the utilities, as will be explained in the following section.

Pyrolysis gasoline (pygas), a byproduct of stream cracking, can be further processed in the BTX complex to recover the aromatic compounds and the raffinate after extraction can be blended in the gasoline or naphtha pool (Balaraman, 2006). If there is no existing aromatics complex to further process the pygas, it could alternatively be routed to the reformer feed for further processing (Philpot, 2007). However, this alternative may not be viable in general as most reformers run on maximum capacity. Pygas from steam cracking contains large amounts of diolefins which are undesirable due to their instability and tendency to polymerize yielding filter plugging compounds. For this reason, hydrogenation of pygas is usually recommended prior to further processing.

1.6.2

Utilities Integration

Petroleum refining and the basic petrochemical industry are the most energy intensive processes in the chemical process industry (Ren, Patel and Blok, 2006). The energy sources in these processes assume different forms including fuel oil, fuel gas, electrical power, and both high and low pressure steam. The different energy requirements and waste from the whole range of refinery and petrochemical units present intriguing opportunities for an integrated complex. Integration of energy sources and sinks of steam cracking, for instance, with other industrial processes, particularly natural gas processing, can yield significant energy savings reaching up to 60% (Ren, Patel and Blok, 2006). Furthermore, gas turbine integration (GTI) between petrochemical units and ammonia plants can lead to a reduction in energy consumption by up to 10% through exhaust-heat recovery (Swaty, 2002). This can be readily extended to the refinery processing units which span a wide variety of distillation, cracking, reforming, and isomerization processes.

Hydrogen is another crucial utility that is receiving more attention recently, mainly due to the stricter environmental regulations on sulfur emissions. Reduction of

sulfur emissions is typically achieved by deeper desulfurization of petroleum fuels which in turn requires additional hydrogen production (Crawford, Bharvani and Chapel, 2002). A less capital intensive alternative to alleviate hydrogen shortage is to operate the catalytic reformer at higher severity. However, higher severity reforming increases the production of BTX aromatics which consequently affect the gasoline pool aromatics specification. Therefore, the BTX extraction process becomes a more viable alternative for the sake of aromatics recovery as well as maintaining the gasoline pool within specification (Crawford, Bharvani and Chapel, 2002). The capital cost for the implementation of such a project would generally be lower as the BTX complex and refinery would share both process and utilities streams.

1.6.3

Fuel Gas Upgrade

Refinery fuel gas is generated from refinery processes and is mainly comprised of C₁/C₂ fractions and some hydrogen. Considerable amounts of light hydrocarbons are produced from the different conversion units in the refinery and are collected in the common fuel gas system. For instance, FCC off gas contains significant amounts of ethylene and propylene which can be extracted and processed as petrochemical feedstocks. A number of integrated U.S. and European refineries have recognized and capitalized on this opportunity by recovering these high value components (Swaty, 2002). This type of synergy requires proper planning and optimization between the petroleum refining and petrochemical complexes.

The other major component is hydrogen which typically accounts for 50–80% of the refinery fuel gas. This substantial amount of hydrogen is passed to the fuel gas system from different sources in the refinery. The most significant source, however, is the catalytic reforming. Hydrogen recovery using economically attractive technologies is of great benefit to both refineries and petrochemical systems, especially with the increasing strict environmental regulations on fuels.

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