Hydroprocessing:
Hydrocracking & Hydrotreating

Chapters 7 & 9
Purpose

Hydrotreating

- Remove hetero atoms & saturate carbon-carbon bonds
  - Sulfur, nitrogen, oxygen, & metals removed
  - Olefinic & aromatic bonds saturated
- Reduce average molecular weight & produce higher yields of fuel products
- Minimal cracking
- Minimal conversion – 10% to 20% typical
- Products suitable for further processing or final blending
  - Reforming, catalytic cracking, hydrocracking

Hydrocracking

- Severe form of hydroprocessing
  - Break carbon-carbon bonds
  - Drastic reduction of molecular weight
- 50%+ conversion
- Products more appropriate for diesel than gasoline
Characteristics of Petroleum Products

Refining Overview – Petroleum Processes & Products,
by Freeman Self, Ed Ekholm, & Keith Bowers, AIChE CD-ROM, 2000
Hydroprocessing Trends

- Hydrogen is ubiquitous in refinery & expected to increase
  - Produces higher yields & upgrade the quality of fuels

- The typical refinery runs at a hydrogen deficit
  - As hydroprocessing becomes more prevalent, this deficit will increase
  - As hydroprocessing progresses in severity, the hydrogen demands increase dramatically

- Driven by several factors
  - Heavier & higher sulfur crudes
  - Reduction in demand for heavy fuel oil
  - Increased use of hydrodesulfurization for low sulfur fuels
  - More complete protection of FCCU catalysts
  - Demand for high quality coke
  - Increased production of diesel
Sources of Hydrogen

- **Catalytic Reformer**
  - *The most important source of hydrogen for the refiner*
  - Continuously regenerated reformer: 90 vol%
  - Semi-continuously regenerated reformer: 80 vol%

- **FCCU Offgas**
  - 5 vol% hydrogen with methane, ethane & propane
  - Several recovery methods (can be combined)
    - Cryogenic
    - Pressure swing adsorption
    - Membrane separation

- **Steam-Methane Reforming**
  - *Most common method of manufacturing hydrogen*
  - 90 to 95 vol% typical purity

- **Synthesis Gas**
  - Gasification of heavy feed
  - Hydrogen recovery – pressure swing adsorption or membrane separation
  - More expensive than steam reforming but can use low quality by product streams
Hydroprocessing Catalysts

Hydrotreating
- Desired function
  - Cobalt molybdenum: sulfur removal & olefin saturation
  - Nickel molybdenum: nitrogen removal & aromatic saturation
- Reactor configuration
  - Fixed bed – temperature to control final sulfur content
- Selective catalysts for sulfur removal without olefin saturation
  - Maintaining high octane rating

Hydrocracking
- Crystalline silica alumina base with a rare earth metal deposited in the lattice
  - Platinum, palladium, tungsten, and/or nickel
- Feed stock must first be hydrotreated
- Catalysts deactivate & coke does form even with hydrogen present
  - Hydrocrackers require periodic regeneration of the fixed bed catalyst systems
  - Channeling caused by coke accumulation a major concern
  - Can create hot spots that can lead to temperature runaways
- Reactor configuration
  - Ebullient beds – pelletized catalyst bed expanded by upflow of fluids
  - Expanded circulating bed – allows continuous withdrawal of catalyst for regeneration
<table>
<thead>
<tr>
<th>Company</th>
<th>State</th>
<th>Site</th>
<th>Atmospheric Crude Distillation Capacity (barrels per stream day)</th>
<th>Desulfurization, Diesel Fuel Downstream Charge Capacity, Current Year (barrels per stream day)</th>
<th>Desulfurization, Gasoline Downstream Charge Capacity, Current Year (barrels per stream day)</th>
<th>Desulfurization, Heavy Gas Oil Downstream Charge Capacity, Current Year (barrels per stream day)</th>
<th>Desulfurization, Kerosene And Jet Downstream Charge Capacity, Current Year (barrels per stream day)</th>
<th>Desulfurization, Naphtha/Reformer Feed Downstream Charge Capacity, Current Year (barrels per stream day)</th>
<th>Desulfurization, Other Distillate Downstream Charge Capacity, Current Year (barrels per stream day)</th>
<th>Desulfurization, Residual Downstream Charge Capacity, Current Year (barrels per stream day)</th>
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# Hydrotreating Technologies

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<th>Provider</th>
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Characteristics of Hydrotreating

![Graph showing hydrogen consumption vs pressure for different processes.](image)

Hydrodesulfurization

- **Sulfur**
  - Sulfur converted to hydrogen sulfide ($H_2S$)
    - Added hydrogen breaks carbon-sulfur bonds & saturates remaining hydrocarbon chains
  - Form of sulfur bonds
    - Sulfur in naphtha generally mercaptans (thiols) & sulfides
    - In heavier feeds, more sulfur as disulphides & thiophenes
  - Light ends
    - Heavier distillates make more light ends from breaking more complex sulfur molecules

- **Unsaturated carbon-carbon bonds**
  - Olefins saturated – one hydrogen molecule added for each double bond
    - Olefins prevalent in cracked streams – coker or visbreaker naphtha, catalytic cracker cycle oil, catalytic cracker gasoline
  - Aromatic rings hydrogenated to cyclopentanes (naphthenes)
    - Severe operation
    - Hydrogen consumption strong function of complexity of the aromatics
    - Prevalent in heavy distillate hydrotreating, gas oil hydrotreating, hydrocracking

- **Selective catalysts available for hydrotreating cat gasoline for sulfur removal but not saturate olefins**
  - Maintain high octane ratings
Hydrogen Consumption

- Chemical consumption due to hydrogenation reactions
  - Cracking reactions of carbon-carbon bonds minimal in hydrotreating, even during aromatic saturation
- Hydrogen is lost in equilibrium with light gases
  - Amount is significant & may double amount required for sulfur removal
- Hydrogen absorbed in liquid products
  - Usually small compared to sulfur removal needs – 1 lb/bbl
- Hydrogen removed with purge gas
  - Used to maintain a high purity of hydrogen — light ends dilute the hydrogen concentration
  - Usually small compared to sulfur removal needs
General Effects of Process Variables

- Reactor inlet temperature & pressure
  - Increasing temperature increases hydrogenation but decreases the number of active catalyst sites
  - Temperature control is used to offset the decline in catalyst activity
  - Increasing pressure increases hydrogen partial pressure & increases the severity of hydrogenation

- Recycle hydrogen
  - Require high concentration of hydrogen at reactor outlet
    - Hydrogen amount is much more than stoichiometric
    - High concentrations required to prevent coke laydown & poisoning of catalyst
      - Particularly true for the heavier distillates containing resins and asphaltenes

- Purge hydrogen
  - Removes light ends & helps maintain high hydrogen concentration
Increasing Severity

- Naphtha hydrotreating
- Distillate (light and heavy) hydrotreating
- Gas oil hydrotreating
Naphtha Hydrotreating

- Naphtha hydrotreated primarily for sulfur removal
  - Mostly mercaptans (RSH) & sulfides (R₂S)
  - Some disulfides (RSSR), & thiophenes (ring structures)
- Cobalt molybdenum on alumina most common catalyst
- Chemical hydrogen consumption typically 50 to 250 scf/bbl
  - For desulfurization containing up to 1 wt% sulfur — 70 to 100 scf/bbl
  - Significant nitrogen & sulfur removal — 250 scf/bbl
Naphtha Hydrotreating Process

- Liquid hourly space velocity ~ 2
- Hydrogen recycle about 2,000 scf/bbl
- Stripper overhead vapor to saturates gas plant
  - Recovery of light hydrocarbons & removal H2S
- Fractionator Pentane/hexane overhead to isomerization
  - Bottoms to reformer

Distillate Hydrotreating

- In general, all liquid distillate streams contain sulfur compounds that must be removed
- Saturate olefins in diesel to improve the cetane number
- Hydrogenation at the high pressure produces small amounts of naphtha from hydrocracking
  - Required to get at the embedded sulfur
  - Diesel hydrotreater stabilizer will have an upper sidestream draw producing the naphtha which is recycled to motor gasoline processing
Distillate Hydrotreating Process

- Hydrogen recycle starts at 2,000 scf/bbl; consumption 100 to 400 scf/bbl
- Conditions highly dependent upon feedstock
  - Distillate (jet fuel & diesel) with 85% - 95% sulfur removal — 300 psig & hydrogen consumption of 200 - 300 scf/bbl
  - Saturation of diesel for cetane number improvement — over 800 scf/bbl hydrogen & up to 1,000 psig

Gas Oil Hydrotreating

- Catalytic cracker feedstocks (atmospheric gas oil, light vacuum gas oil, solvent deasphalting gas oil) hydrotreated severely
  - Sulfur removal
  - Opening of aromatic rings
  - Removal of heavy metals
- Desulfurization of gas oil can be achieved with a relatively modest decomposition of structures
- Gas oils can be contaminated with resins & asphaltenes
  - Deposited in hydrotreater
  - Require catalyst replacement with a shorter run length than determined by deactivation
  - Guard chamber may be installed to prolong bed life
- Nickel molybdenum catalyst system for severe hydrotreating
- Gas oil units more expensive because of more intensive hydrogenation
  - Quench
  - Multi-stage flash
  - More complex strippers
Gas Oil Hydrotreating Process

- Normally two reactor beds – temperature rise
- Hydrogen partial pressure related to ring saturation & amount of sulfur
  - For low ring saturation 300 psig may be sufficient
  - 1,200 psig will convert 25% ring saturation & somewhat less than 95% sulfur removal
  - Pressures as high as 1,500 psig can achieve saturation of 30% of aromatic rings
- Hydrogen absorption of 300 scf/bbl could give about 80% sulfur removal & only require 300 psig
  - No ring saturation at these mild conditions

Hydrocracking

- Purpose: process gas oil to break carbon-carbon bonds of large aromatic compounds & remove contaminants
  - Hydrogenation (addition of hydrogen)
  - Cracking (carbon-carbon scission) of aromatic bonds
- Typically creates distillate range products, not gasoline range products
# U.S. Refinery Hydrocracking

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<th>Company</th>
<th>State</th>
<th>Site</th>
<th>Atmospheric Crude Distillation Capacity (barrels per stream day)</th>
<th>Cat Cracking: Fresh Feed Downstream Charge Capacity, Current Year (barrels per stream day)</th>
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<th>Cat Hydrocracking, Distillate Downstream Charge Capacity, Current Year (barrels per stream day)</th>
<th>Cat Hydrocracking, Gas Oil Downstream Charge Capacity, Current Year (barrels per stream day)</th>
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## Hydrocracking Technologies

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## Use of Yield Charts & Equations

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Hydrocracking Feeds

- Typical feeds
  - Cat cracker “cycle oil”
    - Highly aromatic with sulfur, small ring & polynuclear aromatics, catalyst fines; usually has high viscosity
    - Hydrocracked to form high yields of jet fuel, kerosene, diesel, & heating oil
  - Gas oils from visbreaker
    - Aromatic
  - Gas oil from the delayed coker
    - Aromatic, olefinic, with sulfur

- Usually more economical to route atmospheric & vacuum gas oils to the cat cracker to produce primarily gasoline & some diesel
Gas Oil Hydrocracker Feed

- Hydrocracking does a better job of processing aromatic rings without coking than catalytic cracking
  - Hydrogen used to hydrogenate polynuclear aromatics (PNAs)
  - Reduces frequency of aromatic condensation
- Hydrocracking not as attractive as delayed coking for resid high in resins, asphaltenes & heteroatom compounds
  - Heteroatoms & metals prevalent in resins & asphaltenes poison hydroprocessing catalysts
  - High concentrations of resins & asphaltenes will still ultimately coke
- Feeds limited to a Conradson Carbon Number (CCR) of 8 wt%
- Feeds require high pressures & large amounts of hydrogen
Gas Oil Hydrocracker Products

- Hydrocracking primarily to make distillates
  - In US hydrocracking normally a specialized operation used to optimize catalytic cracker operation
  - In US cat cracking preferred to make gasoline from heavier fractions
- Hydrocracking capacity is only about 8% of the crude distillation capacity
  - Not all refineries have hydrocrackers
- Intent is to minimize the production of heavy fuel oil
  - Light ends are approximately 5% of the feed.
  - Middle distillates (kerosene, jet fuel, diesel, heating oil) still contain uncracked polynuclear aromatics

*All liquid fractions are low in sulfur & olefins*
Hydrocracking Chemistry

- Cracking reactions
  - Saturated paraffins cracked to form lower molecular weight olefins & paraffins
  - Side chains cracked off small ring aromatics (SRA) & cycloparaffins (naphthenes)
  - Side chains cracked off resins & asphaltenes leaving thermally stable polynuclear aromatics (PNAs)
    - But condensation (dehydrogenation) also occurs if not limited by hydrogenation
Hydrocracking Chemistry

Hydrogenation reactions
- Exothermic giving off heat
- Hydrogen inserted to saturate newly formed molecule from aromatic cracking
- Olefins are saturated to form light hydrocarbons, especially butane
- Aromatic rings hydrogenated to cycloparaffins (naphthenes)
- Carbon-carbon bonds cleaved to open aromatic & cycloparaffins (naphthenes) rings
- Heteroatoms form hydrogen sulfide, ammonia, water, hydrogen chloride
Hydrocracking Chemistry

- **Isomerization Reactions**
  - Isomerization provides branching of alkyl groups of paraffins and opening of naphthenic rings

- **Condensation Reactions**
  - Suppressed by hydrogen
Hydrogen Consumption

- Carbon bonds with heteroatoms broken & saturated
  - Creates light ends
    - Heavier distillates make more light ends from breaking more complex molecules
  - Sulfur converted to hydrogen sulfide
  - Nitrogen converted to ammonia
  - Oxygen converted to water
  - Organic chlorides converted to hydrogen chloride
Hydrogen Consumption

- Saturation of carbon-carbon bonds
  - Olefins saturated to form light hydrocarbons.
    - Consumption stoichiometric — one hydrogen molecule added for each double bond
  - Aromatic rings hydrogenated to cycloparaffins (naphthenes).
    - Severe operation — hydrogen consumption strong function of complexity of the aromatics
- Isomerization reactions generally not present
- Metals deposited directly on the catalysts
  - Excess metals reduce catalyst activity & promote dehydrogenation (produces coke & hydrogen)
Hydrogen Consumption

- Have cracking of carbon-carbon bonds
  - Severe operation — hydrogen consumption strong function of complexity of the aromatics

- Hydrogen lost in mixture with products
  - Equilibrium with light gases
    - Significant — may double amount required for sulfur removal
  - Absorbed in liquid products
    - Usually small compared to hydrogen used for sulfur removal
  - Lost with purge gas
Single Stage Hydrocracking

Refining Overview – Petroleum Processes & Products,
by Freeman Self, Ed Ekholm, & Keith Bowers, AIChE CD-ROM, 2000
Severe Two Stage Hydrocracking

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