

Sulphur Recovery Process:-^[1-4]

Major cause of air pollution because of S compounds

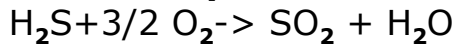
H₂S to S can be converted to S.

S recovery process from acid gas (H₂S).

Claus process :- H₂S oxidation process)

Overall reaction $H_2S + 1/2 O_2 \rightarrow S + H_2O$

In two step:



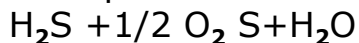
As 22% solution of KOH in water. For maximum sulphur recovery H₂S/SO₂ mole reaction. S recovery efficiency for 3 cascade reactor stage is 96-97% With rich acid gas feed (~50% H₂S) reaction became exothermic where T reactor ~ 1200°C (thermal process)

Clause Process :-

Work well for gas stream containing greater than 20% H₂S (by value) and less than 5% HCS. When reaction is.

1. Heat is recovered from the reaction gases in the waste heat boiler by generating high pressure steam.
2. Reaction gases leaving the waste heat boiler are then passed to a condenser where cooling to 150-190°C result in condensation to elemental S formula.
3. The gases containing unconverted H₂S passed to a reactor containing bed of bauxite cat.
4. Finally waste gases are incinerated to oxidize the last traces of H₂S and elemental S and then vented to atmosphere via stack.

Total S recovery with the subsequent class stages ranges from 94 to 96 percent. Complete conversion of H₂S to SO₂ elemental sulphur is not possible due to thermodynamic chemical equilibrium of class reaction. Selective oxidation of H₂S to sulphur can be thermodynamically complete by reaction:



Series of proprietary catalyst for direct oxidation of H₂S to elemental S are used (Hi activity process).

Sweetening process

Alkyl & aromatic mercaptans :-

Important S compounds distributed in petroleum products. Cause foul odour oxidative determination towards metals. Also mercaptans cause oxidation determination as well as inhibit the performance of various

additives(TEL,antioxidants) in finished products.Removal of is essential (sweetening process).

Mercaptas removal Process

1. Caustic scrubbing, merox extraction.
2. Convation of mercaptous to disulfide include doctor sweetenies ,merox sweetening ,copper chloride sweetening.
3. Acid treating ,clay treating ,catalytic process.

Doctor 2 Sweetening-A

Feed:All naphthas.

mercaptan level :no limits.

Very versatile process.

Disadvantages:Polysulfide formation

Copper Sluxry :^[1-2,5-8]

Feed :Most cracked & straight from naphthas . mercaptans level = up to 0.04%

Doctor Treating Process:

(Also caled sodium plumbite treating) Na_2PbO_2 .

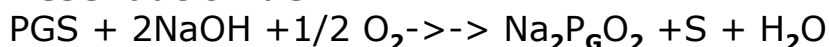
Doctor solution:Active reagent sodium plumbite in aqueous caustic soda.Doctor solution reacts with mercaptans to form lead mercaptides .Sulphur reacts and leads mercaptides and gives disulphide's and lead sulphide.Lead sulphide is regenerated by air , NaOH to sodium plumbite again.

Disadvantage :Difficulty of controlling poly sulphide formation , which reduces octane number,gum content and color stability.

Reaction Involved:

- $\text{P}_6\text{O} + 2\text{NaOH} \rightarrow \text{Na}_2\text{P}_6\text{O}_2 + \text{H}_2\text{O}$
- $2\text{RSH} + \text{Na}_2\text{P}_6\text{O}_2 \rightarrow (\text{RS})_2\text{P} + 2\text{NaOH}$
- $(\text{RS})_2\text{P}_6 + \text{S} \rightarrow \text{R}_2\text{S}_2 + \text{P}_6\text{S}$

Aesenuation def:



Coppa Chloride Sweetening

Used to sweeter gasoline and kerosene by direct oxidation of mercaptans to disulfides by direct oxidation of mercaptans to disulfides by using cupric chloride oxidation as oxidizing agents.

Fundamental reaction invalid are:

- $2\text{RSH} + \text{CuCl}_2 \rightarrow (\text{RS})_2\text{Cu} + 2\text{HCL}$.
- $(\text{RS})_2\text{Cu} + \text{CuCl} \rightarrow \text{RSSR} + \text{Cu}_2\text{Cl}_2$
- $\text{Cu}_2\text{Cl}_2 + 2\text{HCL} \rightarrow \text{C}_{42}\text{Cl}_2 + 2\text{HCL}$
- $\text{Cu}_2\text{Cl}_2 + 2\text{HCL} + \frac{1}{2}\text{O}_2 \rightarrow 2\text{C}_4\text{Cl}_2 + \text{H}_2\text{O}$

Water produced must be removed continuously to maintain correct reaction condition.

Advantage:

No loss of octane no.

Alternative source of Energy^[3-4]

- ▶ Due to world population growth and the advance of technologies that depend on fossil fuels, reserves of fossil fuel eventually will not be able to meet energy demand.
- ▶ Energy experts point out that reserves are less than 40 years for petroleum, 60 years for natural gas and 250 years for coal.
- ▶ Fossil fuel costs are likely to increase in the near future. This will allow renewable energy sources such as solar, wind, hydrogen, etc., to be utilized.

Ethanol Production From Sugar Crops

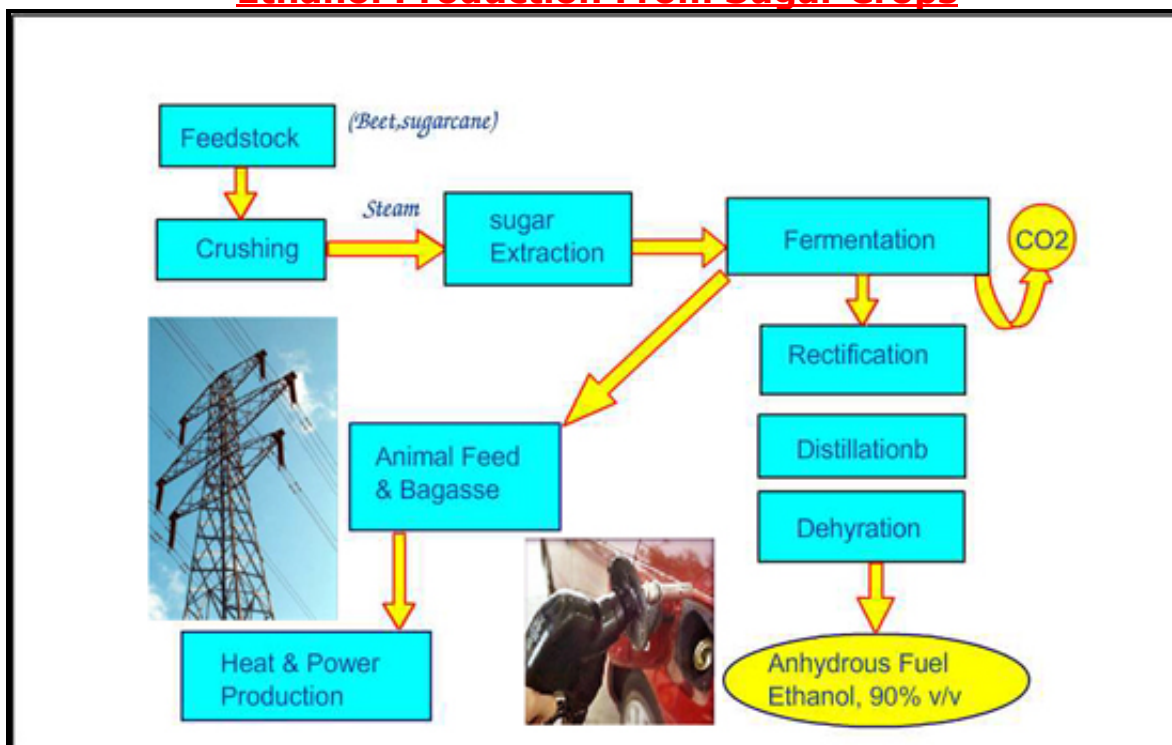


Fig:9.1 Ethanol Production From Sugar Crops^[4-9]

Table:9.1

Properties Of Conventional and Alcohol Fuels

| Characterstics | Diesel | Gasoline | Methanol | Ethanol |
|-----------------------|---------|----------|----------|---------|
| Energy Content(MJ/Kg) | 42.5 | 44.0 | 20.0 | 26.9 |
| Bioling Point,°C | 140-360 | 37-205 | 65 | 78 |
| Reaserch Octane No | - | 87-98 | 106 | 107 |
| Motor Octane No | - | 80-90 | 92 | 89 |
| Cetane No | 45-55 | 0-5 | 5 | 5 |

Emissions with Ethanol Gasoline Blends

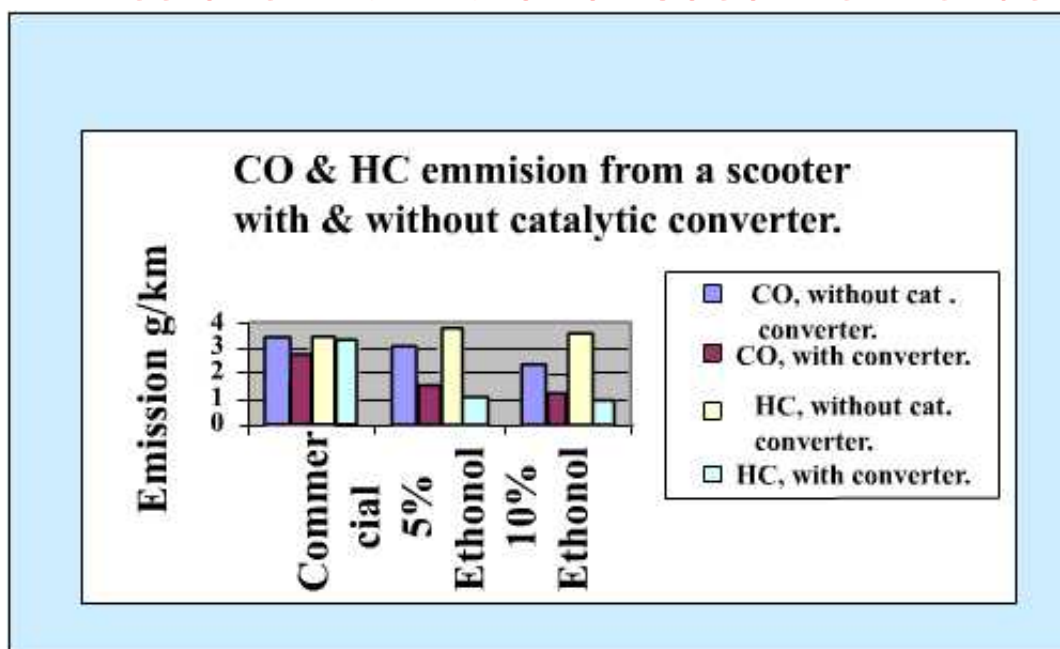


Fig:9.2 Emissions with Ethanol Gasoline Blends^[8,9]

Ethanol as Transport Fuel^[3-7,9-14]

Ethanol Advantage as Transport Fuel

- ▶ Octane Enhancer
- ▶ No engine modification required for E₁₀ and lower blend
- ▶ Higher compression operation of the engine feasible

Environmental Benefits

- ▶ Lower emission of CO, VOC and Hydrocarbons
- ▶ Lower CO₂ emission with higher compression ratio

- ▶ Complete CO₂ cycle
- ▶ Higher biodegradability and low toxicity

5% Ethanol blending in gasoline is mandatory in India. Further, BIS is examining the issues related to E₁₀ blending.

CO₂ Cycle of Ethanol Production From Sugarcane

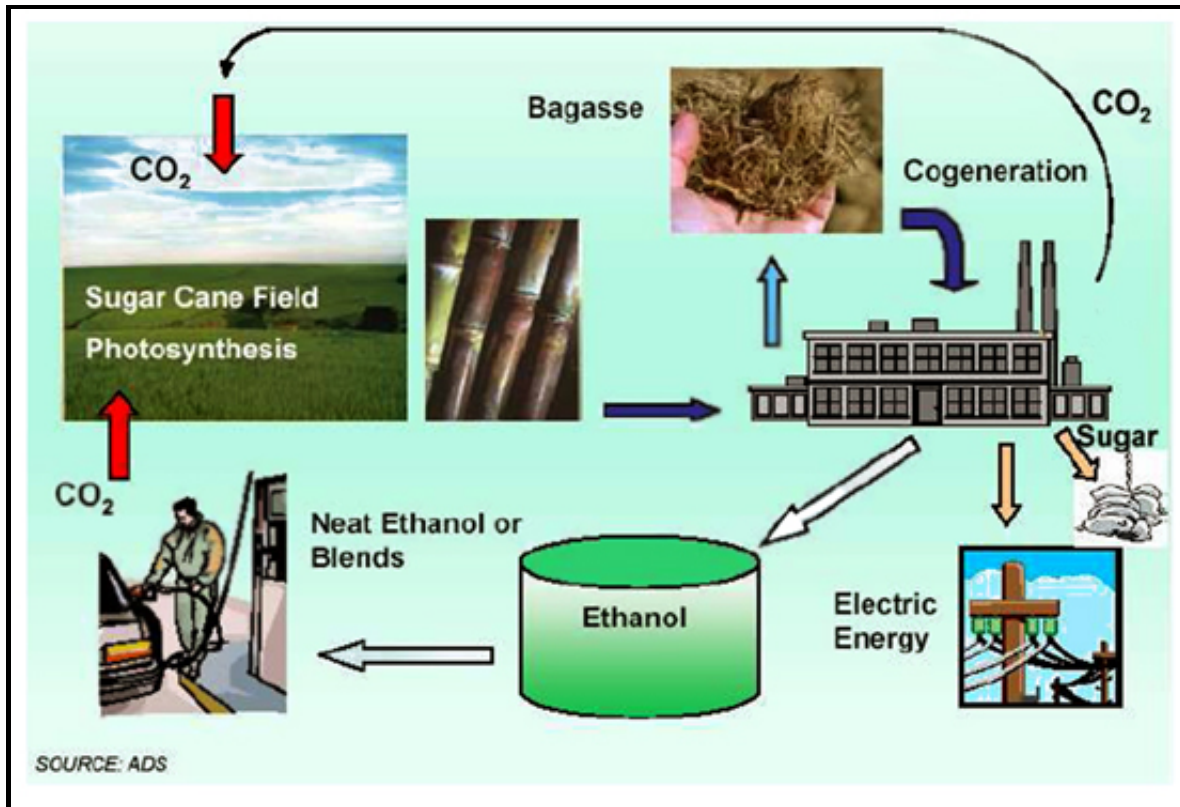


Fig:9.3 Cycle of Ethanol Production From Sugarcane

Observation of Physico-Chemical Properties of Blend

- ▶ Octane number of gasoline increases with ethanol blending which offer opportunities to refineries for reducing benzene and MTBE.
- ▶ Copper impurity in the anhydrous ethanol act as precursor for oxidation of the blend.
- ▶ Additional dosage of anti-oxidants will be required to meet potential gum specification and engine deposit formation.
- ▶ Addition of corrosion inhibitor required as blend have been reported to cause problem on carburetor needles of two wheelers.

BIO-DIESEL [7-9]



Biodiesel is a monoalkylester of long chain fatty acids produced from the trans-esterification reaction of vegetables oils with alcohol in presence of catalyst and can be used as a fuel.

Fig:9.4

BIODIESEL-Fuel From Oil Crops

Feed Stocks

- Rapeseed Oil (Europe)
- Sunflower Oil (Italy & France)
- Soybean Oil (USA & Brazil)
- Palm Oil (Malaysia)
- Linseed, Olive Oil (Spain)
- Jatropha & Karanjia (India)

Production Process: Transesterification Path

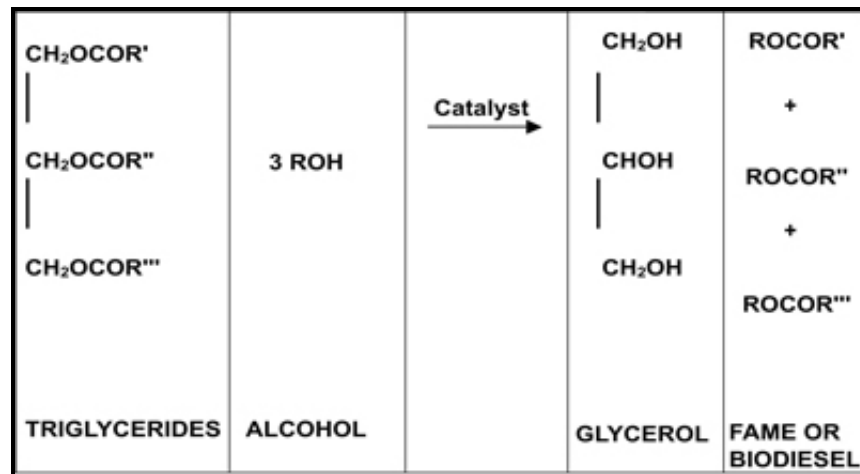


Fig:9.5

- High Cetane
- Better Lubricity
- No Sulphur
- Can be directly used in diesel engine
- Environment Friendly
-

Complete CO Cycle

2

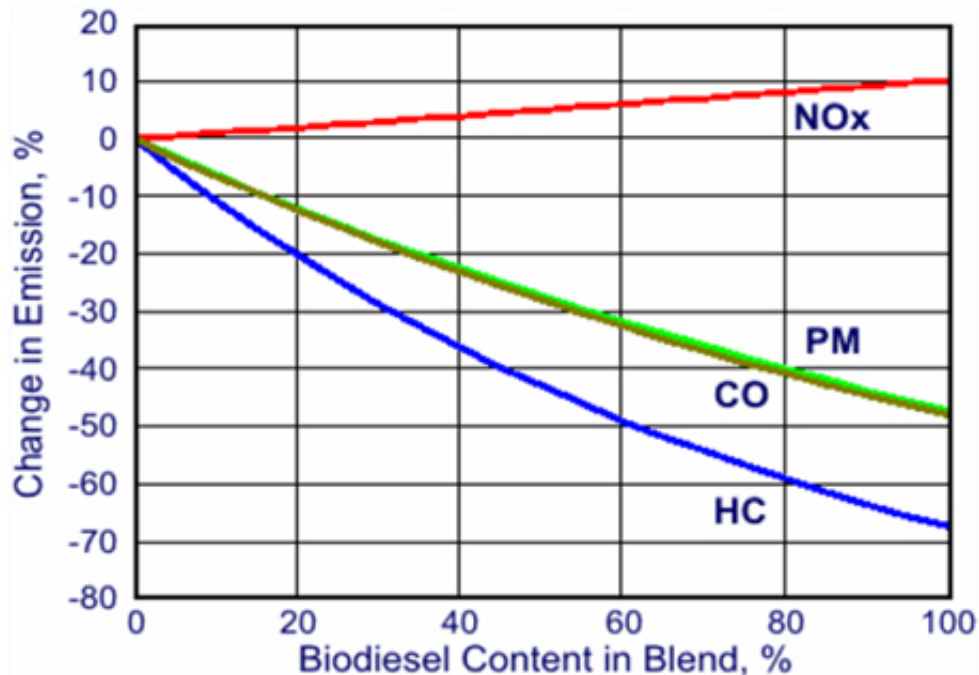
Table:9.2 Transesterification Reaction for Biodiesel Production^[8,9]**Wide Global Acceptance of Biodiesel**

- ▶ Zero Emissions
- ▶ By leading automobile manufacturers Audi, BMW, Kubota, Massey-fergusan, Mercedes-Benz, Nissan Peugeot, Renault, Skoda, Volkswagen, Volvo etc.
- ▶ By the Fuel Trade: Chevron-Texaco, Shell, TOTAL, Exxaon Mobile, BP etc
- ▶ By end user enterprises: Bus companies, taxi-fleets, forestry enterprises, railroad, boat owners etc

Raw Material for Biodiesel Production

- ▶ Rapeseed Oil (Europe)
- ▶ Sunflower Oil (Italy & France)
- ▶ Soybean Oil (USA & Brazil)
- ▶ Palm Oil (Malaysia)
- ▶ Linseed, Olive Oil (Spain)
- ▶ CottonSeeds oil (Greece)
- ▶ Jatropha & Pongamia (Karanja) in India
- ▶ Jatropha (Nicaragua & SouthAmerica)

Impact of Bio-diesel on Emissions



Ref : NREL, US DOE

Fig:9.6 Impact of Bio-diesel on Emissions

2nd Generation Biofuels^[3-6]

- ▶ 2nd generation biofuels have high emissions reduction potential.
- ▶ Biomass to be used include woody crops and grasses(not competitive with food crops).
- ▶ High yield/Hectare as compare to first generation biofuels (BTL>3100 L/HC oo HSD equivalent)
- ▶ BTL result in designer fuels compatible with new technologies
- ▶ Technology option for 2nd generation fuels:
 - ▶ Direct fired combustion
 - ▶ Gasification
 - ▶ Pyrolysis
 - ▶ Cofiring

Various Biomass Conversion Technologies

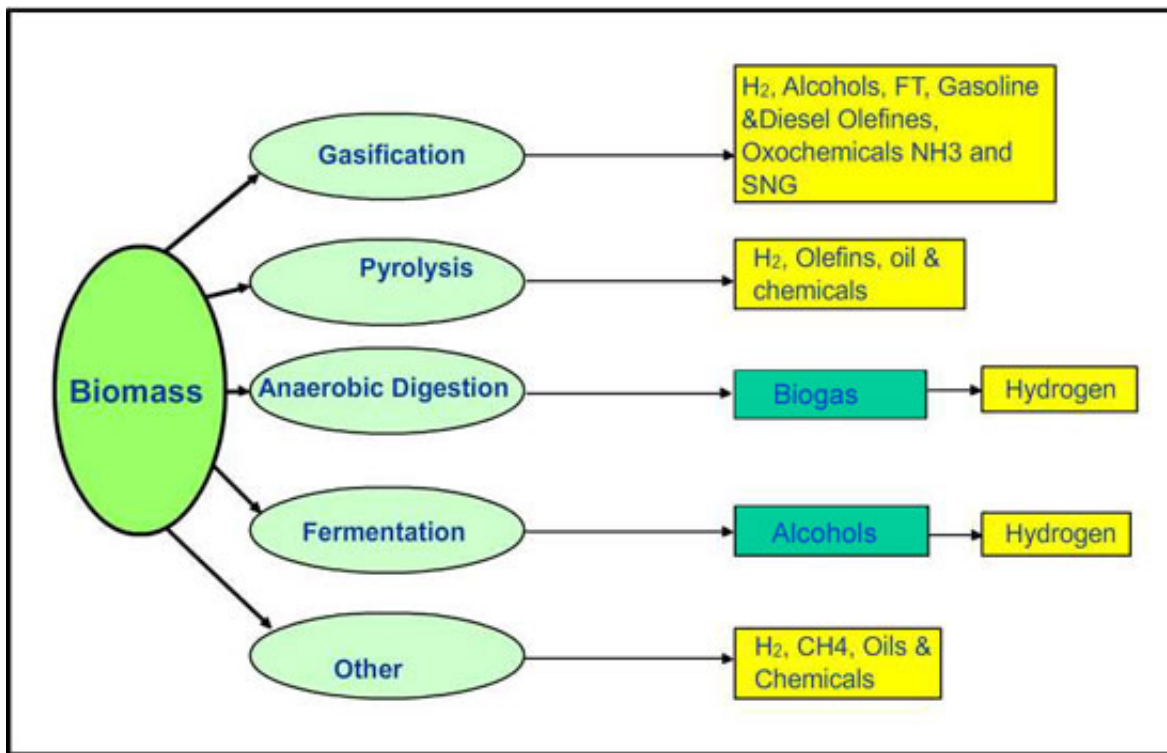


Fig:9.7 Various Biomass Conversion Technologies

Hydrogen as Future Fuel

- ▶ Available from water using non petroleum fossil fuel or renewable energy resource.
- ▶ Recycle rapidly and cleanly from hydrogen to water.
- ▶ Clean burning-potential environmental benefits.
- ▶ Provides High efficiency energy conversion process
- ▶ Compatible with all energy uses.

Table:9.3

| Comparison of key properties for hydrogen and other fuels | | | | |
|--|-----------------------------------|--|------------------------|---|
| Fuel type | Energy Per Unit Mass(j/kg) | Energy Per Unit Volume(j/m³) | Motivity Factor | Specific Carbon Emission(kg C/kg fuel) |
| Liquid Hydrogen | 141.90 | 10.10 | 1.00 | 0.00 |
| Gaseous Hydrogen | 141.90 | 0.013 | 1.00 | 0.00 |
| Fuel Oil | 45.50 | 38.65 | 0.78 | 0.84 |
| Gasoline | 47.50 | 34.85 | 0.76 | 0.86 |

| | | | | |
|--------------------|--------------|--------------|-------------|-------------|
| Jet Fuel | 46.50 | 35.30 | 0.75 | - |
| LPG | 48.80 | 24.40 | 0.62 | - |
| LNG | 50.00 | 23.00 | 0.61 | - |
| Methanol | 22.30 | 18.10 | 0.23 | 0.50 |
| Ethanol | 29.90 | 23.60 | 0.37 | 0.50 |
| Bio diesel | 37.00 | 33.00 | - | 0.50 |
| Natural gas | 50.00 | 0.04 | 0.75 | 0.46 |
| Charcol | 30.00 | - | - | 0.50 |

Current & Future Hydrogen Production Options^[8-9]

Thermal Processes

- Reforming of Natural Gas/Naptha
- Gasification of Extra Heavy Oil /Coal/Biomass
- High-Temperature Water Splitting

Eletrolytic Processes

- Electricity from renewable sources like wind,solar,hydel etc.

Photolytic Processes

- Photobiological Water Splitting
- Photoelectrochemical Water Splitting

Other Options

- Chlor-Alkali Plants
- Co-generation electricity from Bagasse at sugar mills

H₂ PRODUCTION-GASIFICATION OPTION

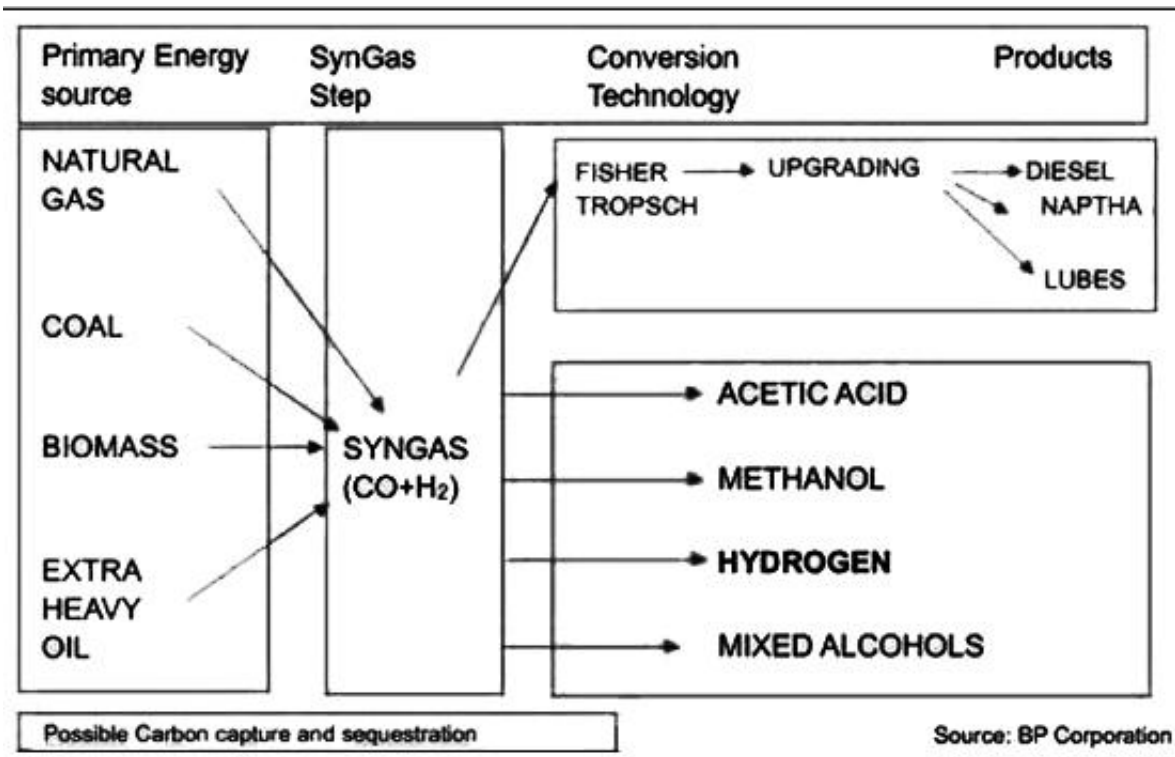


Fig:9.8 H₂ PRODUCTION-GASIFICATION OPTION

Fuel Cells^[18-19]

- ▶ Fuel cell is essentially a battery that does not need recharging. As long as hydrogen and oxygen fuel are supplied, it can continue to supply heat and an electrical current indefinitely.
- ▶ Fuel cell consists of an electrolyte(a conductor of charged particles) between an anode(negatively charged electrode)and a cathode (a positively charged electrode).
- ▶ Once activated by a catalyst, the hydrogen gas separates into protons and electrons, and the electrons are conducted through a wire, forming an electrical current.
- ▶ The protons move through the electrolyte, where they combine with oxygen and other electrons to produce heat and a water by product.

FUEL CELL^[18-22]

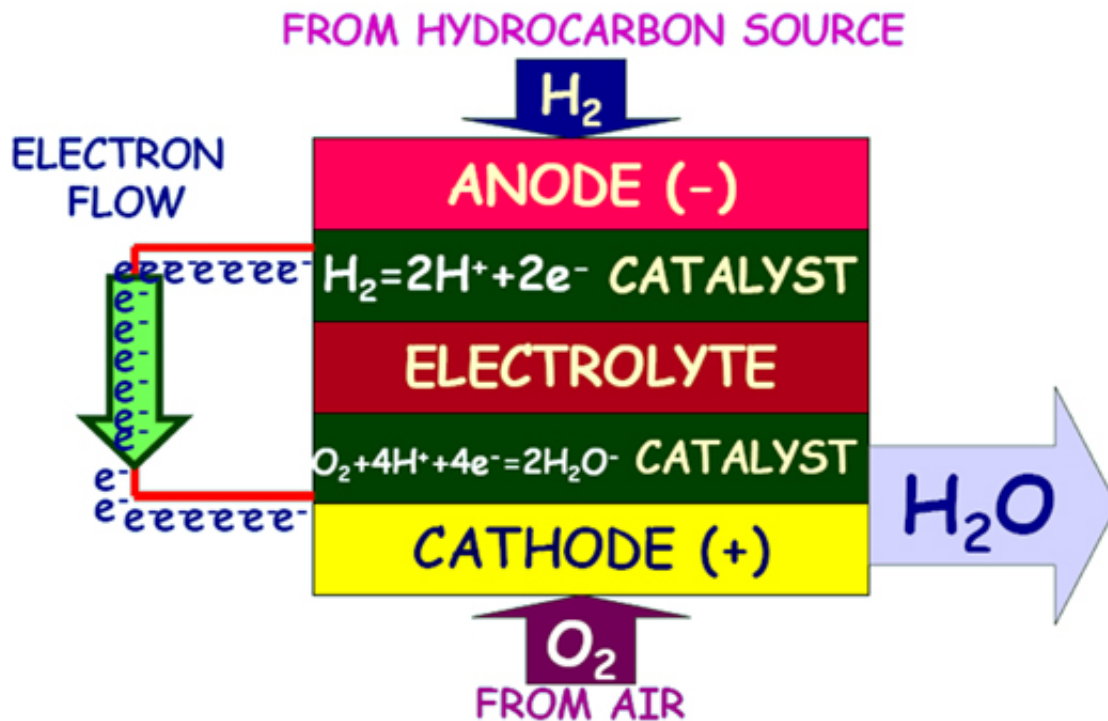


Fig:9.9 Fuel Cell

Fuel Cell Benefits^[18-22]

- Zero Emissions
- High Efficiency
- Environmentally Benign
- Quiet and Quick and Easy Refueling
- Wide Range of Operating Temperatures
- Minimal Maintenance
- Reliability and Quick Startup
- Load Following
- Ultimate Solution for Hydrogen Commercialization

Pollution control in Refining^[22-24]

A petroleum refinery is generally considered as a single energy object, the energy efficiency of which is approximately 17-20%. This is primarily due to the inadequate utilization of secondary energy resources in the process units (30-35%) and the almost complete loss of low-potential heat. To do enhance the energy recovery, energy-technological schemes must be introduced more widely, process units must be fitted with boiler utilizers, the energy from liquid and vapor streams must be used to produce electric power, and the efficiencies of heat exchange equipment must be increased. A considerable fuel saving can be achieved by the use of low-potential heat in heat pumps. Possibilities are available for the reduction of discharges of sulfur dioxide, hydrogen sulfide, and other sulfur compounds that pollute the biosphere. Coming into petroleum refineries every year along with the crude oil is up to five million metric tons of sulfur, which is a raw material in short supply for the production of sulfuric acid. However, thus far no more than 10% of

this sulfur has been recovered from petroleum. The remaining sulfur either passes into the petroleum products, is partly burned in flares with the acid gases, is discharged to the atmosphere, or is dissolved in the wastewater. On the average, in the combustion of medium-sulfur fuel in tube furnaces, the combustion of product vapors in flares, and the burnoff of coke in catalytic cracking units, the processing of one metric ton of crude oil results in the discharge of up to 14 kg of sulfur dioxide and 0.23 kg of hydrogen sulfide to the atmosphere.

Sources of waste water generated from Refineries^[22-25]

Refinery waste waters can be characterized as:-

- Cooling Towers
- Boiler Feed water treatment units
- Process Units
- Desalter brine
- Tank Farms
- Chemical waste
- Sanitary and miscellaneous use
- Fire protection

The capacity of the environment to absorb the effluents and other impacts of process technologies is not unlimited, as some would have us believe. The environment should be considered to be an extremely limited resource, and discharge of chemicals into it should be subject to severe constraints. Indeed, the declining quality of raw materials, especially petroleum and fossil fuels that give rise to many of the gaseous emissions of interest in this text, dictates that more material must be processed to provide the needed fuels .

Oil from subterranean and submarine reservoir extraction can be environmentally damaging. Crude oil and refined fuel spills from tanker ship accidents have damaged fragile ecosystems. Burning oil releases carbon dioxide into the atmosphere, which contributes to global warming. Thus, both the production and processing of crude oil involve the use of a variety of substances, some toxic, including lubricants in oil wells and catalysts and other chemicals in refining. The amounts used, however, tend to be small and relatively easy to control. More detrimental to the environment is the spillage of oil, which has been a particularly common event.

Minor losses from truck and car accidents can affect rivers and streams. Leakage from underground gasoline storage tanks, many abandoned decades ago , has contaminated some local water supplies and usually requires expensive operations either to clean or seal off. Both the production and processing of crude oil involve the use of a variety of substances, some toxic, including lubricants in oil wells and catalysts and other chemicals also in refining. The amounts used, however, tend to be small and relatively easy to control. More detrimental to the environment is the spillage of oil, which has been a particularly common event.

Minor losses from truck and car accidents can affect rivers and streams. A pollutant is a substance present in a particular location (ecosystem) when it is not indigenous to the location or is present in a greater-than-natural concentration. The substance is often the product of human activity.

Pollutants are subdivided into two classes: primary and secondary.

Source -> Primary pollutant -> Secondary pollutant

GASEOUS EMISSIONS^[24-26]

Gaseous emissions from petroleum refining create a number of environmental problems. During combustion, the combination of hydrocarbons, nitrogen oxide, and sunlight results in localized low-levels of ozone, or smog. This is particularly evident in large urban areas and especially when air does not circulate well. Petroleum use in automobiles also contributes to the problem in many areas. The primary effects are on the health of those exposed to the ozone, but plant life has been observed to suffer as well. Refinery and natural gas streams may contain large amounts of acid gases, such as hydrogen sulfide (H₂S) and carbon dioxide (CO₂).

Hydrogen chloride (HCl), although not usually considered to be a major pollutant in petroleum refineries can arise during processing from the presence of brine in petroleum that is incompletely dried. It can also be produced from mineral matter and other inorganic contaminants and is gaining increasing recognition as a pollutant which needs serious attention. Acid gases corrode refining equipment, harm catalysts, pollute the atmosphere, and prevent the use of hydrocarbon components in petrochemical manufacture. When the amount of hydrogen sulfide is large, it may be removed from a gas stream and converted to sulfur or sulfuric acid. Some natural gases contain sufficient carbon dioxide to warrant recovery as dry ice, that is, solid carbon dioxide. And there is now a conscientious effort to mitigate the emission of pollutants from hydrotreating process by careful selection of process parameters and catalysts.

The terms refinery gas and process gas are also often used to include all of the gaseous products and by-products that emanate from a variety of refinery processes. There are also components of the gaseous products that must be removed prior to release of the gases to the atmosphere or prior to use of the gas in another part of the refinery, that is, as a fuel gas or as a process feedstock. In addition to the gases obtained by distillation of petroleum, more highly volatile products result from the subsequent processing of naphtha and middle distillate to produce gasoline. Hydrogen sulfide is produced in the desulfurization processes involving hydrogen treatment of naphtha, distillate, and residual fuel; and from the coking or similar thermal treatments of vacuum gas oils and residual fuels. The most common processing step in the production of gasoline is the catalytic reforming of hydrocarbon fractions in the heptane (C₇) to decane (C₁₀) range.

Natural gas is also capable of producing emissions that are detrimental to the

environment. While the major constituent of natural gas is methane, there are components such as carbon dioxide (CO), hydrogen sulfide (H₂S), and mercaptans (thiols; R-SH), as well as trace amounts of sundry other emissions. The fact that methane has a foreseen and valuable end-use makes it a desirable product, but in several other situations it is considered a pollutant, having been identified a greenhouse gas.

In addition to the corrosion of equipment of acid gases, the escape into the atmosphere of sulphur containing gases can eventually lead to the formation of the constituents of acid rain, that is, the oxides of sulfur (SO₂ and SO₃). Similarly, the nitrogen-containing gases can also lead to nitrous and nitric acids (through the formation of the oxides NO_x,) which are the other major contributors to acid rain. The release of carbon dioxide and hydrocarbons as constituents of refinery effluents can also influence the behavior and integrity of the ozone layer. Gases such as sulfur oxides (usually sulfur dioxide, SO₂) as well as nitrogen oxides (NO_x) react with the water in the atmosphere to form acids. Acid rain has a pH less than 5.0 and predominantly consists of sulfuric acid (H₂SO₄) and nitric acid (HNO₃). As a point of reference, in the absence of anthropogenic pollution sources the average pH of rain is 6.0 (slightly acidic; neutral pH 7.0).

In summary, the sulfur dioxide that is produced during a variety of processes will react with oxygen and water in the atmosphere to yield environmentally detrimental sulfuric acid. Similarly, nitrogen oxides will also react to produce nitric acid. Hydrogen chloride, if produced during refining, quickly picks up moisture in the atmosphere to form droplets of hydrochloric acid and, like sulfur dioxide, is a contributor to acid rain. However, hydrogen chloride may exert severe local effects because, unlike sulphur dioxide, it does not need to participate in any further chemical reaction to become an acid and under atmospheric conditions that favor a buildup of stack emissions in the area of a large industrial complex or power plant, the amount of hydrochloric acid in rainwater could be quite high.

Liquid Effluents

Crude oil, as a mixture of hydrocarbons, is (theoretically) a biodegradable material. However, in very general terms, petroleum is a mixture of: (a) hydrocarbons, (b) nitrogen compounds, (c) oxygen compounds, (d) sulfur compounds, and (e) metallic constituents. It is convenient to divide the hydrocarbon components of petroleum into the following three classes: Crude oil also contains appreciable amounts of organic no hydrocarbon constituents, mainly sulfur, nitrogen, and oxygen-containing compounds and, in smaller amounts, organometallic compounds in solution and inorganic salts in colloidal suspension. These constituents appear throughout the entire boiling range of the crude oil but tend to concentrate mainly in the heavier fractions and in the non-volatile residues.

Air Quality Standards:

They are the standards that specify allowable emission.

It can either be related to:

1. Quality of the air in the surrounding atmosphere (ambient air quality) – Useful for the protection of the health of living populations, animals and of property.
2. Gaseous emissions from a particular unit (emission standard) - used for planning pollution control strategy.

Ambient Air Quality Standards:

In India, for AAQS, various areas have been classified into 3 categories:

1. **Category A:** covers Industrial and mixed – use areas.
2. **Category B:** Covers residential and Rural areas.
3. **Category C:** Covers sensitive areas such as sanctuaries, national parks, hill stations etc.

Table: 9.4 Ambient Air Quality Standards: India

| Category/ Area | Concentration in micrograms/ m ³ | | | |
|-------------------------------------|---|-----------------|-----------------|------|
| | SPM | SO ₂ | NO _x | CO |
| A.Industrial & Mixed Use | 500 | 120 | 120 | 2000 |
| B.Residential & Rural | 200 | 80 | 80 | 1000 |
| C.Sensitive | 100 | 30 | 30 | 500 |

Supporting Processes

- ▶ Not directly involved in the processing of petroleum based fuels
- ▶ Processes
 - Hydrogen production and purification
 - Gas processing units
 - Acid gas treating
 - Sulfur recovery
 - Water treatment

Hydrogen Production & Purification

- ▶ Sources of hydrogen
 - Catalytic Reformer

Majority of hydrogen used in refinery

- FCCU Off Gas
- Steam-Methane Reforming

Most common method of manufacturing hydrogen Methane, ethane, or heavy components reformed to hydrogen, carbon dioxide, & water in a series of three reactions .Hydrogen purity typically 90 to 95 vol%

- Synthesis Gas

Chemical solvent processes

- ▶ Amine sweetening (MEA, DEA, MDEA, DGA)
- ▶ Hot potassium carbonate
 - Physical solvent processes
 - ▶ Selexol
 - ▶ Propylene carbonate
 - ▶ Sulfinol
 - ▶ Rectisol
 - Dry absorbents
 - ▶ Molecular sieve
 - ▶ Activated charcoal
 - ▶ Iron sponge
 - ▶ Zinc Oxide

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