

**IEA Bioenergy Agreement
Task 33: Thermal Gasification of Biomass (2001-2003)**

Technology Brief

**Biomass Gasification to produce Synthesis Gas for Fuel Cells, Liquid Fuels
and Chemicals**

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Introduction and Background

The aim of this work is to give an overview about biomass gasification to produce synthesis gas for fuel cells, liquid fuels and chemicals.

Synthesis Gas which consists mainly of hydrogen and carbon monoxide is a raw material for large scale synthesis for the production of important basis products of the organic chemistry. The fundamental reactions of synthesis gas chemistry are methanol synthesis, Fischer-Tropsch synthesis, oxo synthesis (hydroformylation), and methane synthesis. Also hydrogen and carbon monoxide are produced from synthesis gas. At the moment synthesis gas is produced from natural gas and oil. In the frame of the shortness of oil also the production from coal becomes more important.

To produce synthesis gas from a solid fuel the following steps are necessary:

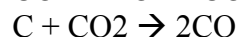
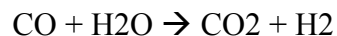
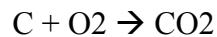
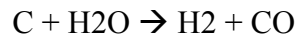
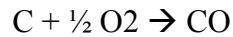
- 1) Gasification of the fuel
- 2) Cleaning of the product gas
- 3) Usage of the synthesis gas (chemical reaction or usage as energy carrier in fuel cells)

Gasification Systems

Gasification is a thermochemical conversion of a solid fuel to a gaseous fuel. The solid fuel reacts with the gasification agent (air, oxygen, steam, carbon dioxide or mixtures of these) to a burnable gas (the main burnable components are hydrogen, carbon monoxide and methane).

The main reactions are:

Devolatilisation of the solid fuel



This reaction can be done in fixed bed, in fluidised bed or in entrained flow reactors.

To produce synthesis gas from biomass a gasification systems is necessary, which produces an almost nitrogen-free product gas. This can be realised by allothermal gasification (to introduce the heat, which is necessary for the gasification reactions from outside of the reactor) or to use oxygen as gasification agent.

The following gasification systems, which are able to produce synthesis gas, are at the moment under development or in demonstration phase:

Allothermal with heat transporting media:

FICFB-Gasification system	http://www.ficfb.at	8MW _{th}
Silvagas gasification system	http://www.future-energy.com/	40MW _{th}
Staged reforming	http://www.dm1-2.de/	1MW _{th}

Allothermal with heat exchangers

Pulse combustor	http://www.electro-farming.de/
Heat pipe reformer	http://www.ltk.mw.tu-muenchen.de/

Oxygen blown gasification systems

Carbo-V Plant	http://www.choren.de/	1MW _{th}
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A detailed description of these gasification systems will be done in Subtask “Circulating Fluidized Bed (CFB) and fluidized bed (FB) Gasification, Gas Cleaning, and Fuel Gas Utilization Systems“ by Esa Kurkela/Pekka Simell, VTT, Finland.

Gas Treatment

Each synthesis gas production plant requires gas treatment facilities to purify product gases and also, in many cases, to condition them. Whereas the gas purification system eliminates the components that would affect downstream processing or utilization of these gases, the purpose of a conditioning system is to adjust gas components to the appropriate ratio. The ratio of hydrogen to carbon monoxide, especially, must be matched to the requirements of downstream synthesis or reduction units and other gas consumers.

The task of gas purification and conditioning is most complex downstream of plants producing synthesis gas from solid fuels. Capital investment in these purification units can be much higher than that required for the gasification plant itself. Treatment of gases produced through gasification of high molecular mass liquid hydrocarbons (tars) is also rather expensive. Generally, the smaller the molecules from which a gas is produced, the lower is the capital investment required in treatment facilities. The costs of gas purification are also strongly dependent on the amount and type of impurities in the feedstock, particularly its content of sulphur and trace elements.

Depending on the type and composition of feedstock and the type of gasification process used, a gas purification plant must handle the following impurities:

- Dust
- Carbonization products, tar, gas liquor
- Carbon
- Cyanic compounds
- Nitrogen oxides
- Carbon dioxide
- Hydrogen sulphide, carbonyl sulphide, organic sulphur compounds
- Ammonia

Other gas components, even if they occur only in trace amounts, require specific measures to prevent long-range damage or malfunction of the plant. Such components may include volatile metal compounds and high molecular mass organic compounds.

There are many different technologies to clean the synthesis gas. They can be divided by the operating temperature: High temperature gas cleaning (above 200°C) and low temperature gas cleaning (below 200°C).

A good overview about high temperature gas cleaning is given in the report: Hot Gas conditioning: Recent Progress with Larger-Scale Biomass Gasification Systems D. J. Stevens, Pacific Northwest National Laboratory, U.S.A.

In the area of low temperature gas cleaning much research was done in the last years. There are many different gas treatment systems on the market and described in literature.

Utilisation of Synthesis Gas in Chemical Industry

3.1 Methanol Synthesis.

Methanol can be produced from hydrogen – carbon oxide mixtures by means of the catalytic reaction of carbon monoxide and some carbon dioxide with hydrogen.



The presence of a certain amount of carbon dioxide in the percentage range is necessary to optimise the reaction. Side reactions, also strongly exothermic, can lead to formation of byproducts such as methane, higher alcohols, or dimethylether.

2520 m³ (STP) of synthesis gas (70 % H₂, 21 % CO, 7 % CO₂) are necessary to produce a tonne of methanol. The purity of the latter depends on the catalyst used. Copper – zinc catalysts used in the low-pressure process require a sulphur-free gas (H₂S <1 ppm [mL/m³]). Catalysts for the high- and medium-pressure processes (ZnO activated with chromic acid) can accept 30 ppm (mL/m³) of hydrogen sulphide.

3.1.1 Process Technology

The oldest process for the industrial production of methanol is the dry distillation of wood, but this no longer has practical importance. Other processes, such as the oxidation of hydrocarbons and production as a byproduct of the Fischer – Tropsch synthesis according to the Synthol process, have no importance today.

Methanol is currently produced on an industrial scale exclusively by catalytic conversion of synthesis gas. Processes are classified according to the pressure used:

- 1) High-pressure process 25 – 30 MPa
- 2) Medium-pressure process 10 – 25 MPa
- 3) Low-pressure process 5 – 10 MPa

The main advantages of the low-pressure process are lower investment and production costs, improved operational reliability, and greater flexibility in the choice of plant size.

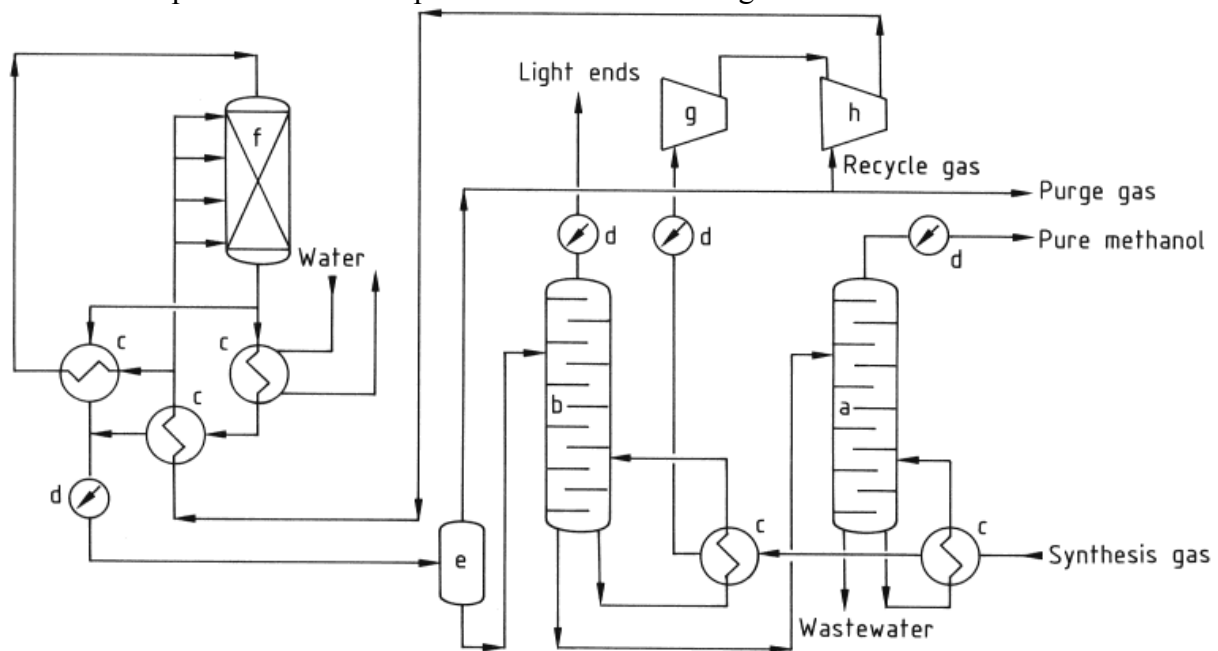
3.1.2 Reactor Design

Current industrial processes for producing methanol differ primarily in reactor design. Many different reactors are available. They may be either adiabatic (e.g., ICI) or quasi-isothermal (e.g., Lurgi). The ICI process accounts for 60 %, and the Lurgi process for 30 % of worldwide methanol production.

Adiabatic Reactors.

The ICI process uses an adiabatic reactor with a single catalyst bed. The reaction is quenched by adding cold gas at several points. Thus, the temperature profile along the axis of the reactor has a sawtooth shape.

The ICI low-pressure methanol process is shown in the figure below:



a) Pure methanol column; b) Light ends column; c) Heat exchanger; d) Cooler; e) Separator; f) Reactor; g) Compressor; h) Compressor recycle stage

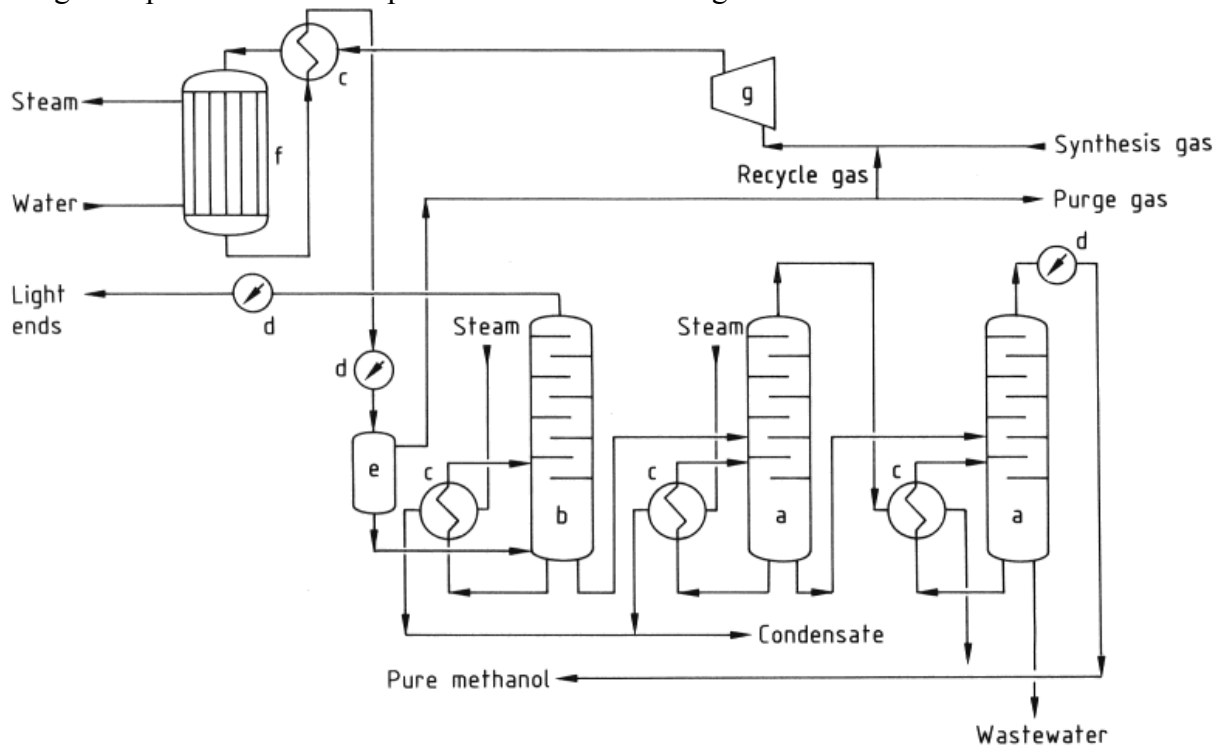
In the Kellogg process, synthesis gas flows through several reactor beds that are arranged axially in series. In contrast to the ICI quench reactor, the heat of reaction is removed by intermediate coolers. The Haldor Topsoe reactor operates on a similar principle, but synthesis gas flows radially through the catalyst beds.

Ammonia – Casale S. A. has developed a reactor that employs a combination of axial and radial flow (mixed flow). This type of reactor initially developed for ammonia plants is offered by Davy McKee in ICI license.

Quasi-Isothermal Reactors.

The Lurgi process employs a tubular reactor (f) with cooling by boiling water. The catalyst is located in tubes over which water flows. The temperature of the cooling medium is adjusted by a preset pressure.

Lurgi low-pressure methanol process is shown in the figure below:



a) Pure methanol columns; b) Light ends column; c) Heat exchanger; d) Cooler; e) Separator; f) Reactor; g) Compressor recycle stage

The Variobar reactor developed by Linde consists of a shell-and-tube reactor coiled in several tiers, whose cooling tubes are embedded in the catalyst packing. The reactor temperature is adjusted by water cooling. As in other processes, the heat of reaction is utilized to produce steam, which can be used, for example, to drive a turbine for the compressor or as an energy source for subsequent methanol distillation.

Whereas synthesis gas flows axially through the two above-mentioned reactors, Toyo offers a reactor through which it flows radially. The advantages, as in the Variobar reactor, lie in a high heat transfer rate with only slight pressure loss.

The Mitsubishi Gas Chemical (MGC) process uses a reactor with double-walled tubes that are filled in the annular space with catalyst. The synthesis gas first flows through the inner tube to heat it up and then, in countercurrent, through the catalyst between the two tubes. The outer tubes are cooled by water, Mitsubishi considers the main advantage of this process to be the high conversion rate (ca. 14 % methanol in the reactor outlet).

3.1.3 Catalysts

3.1.3.1 Catalysts for High-Pressure Synthesis

The first industrial production of methanol from synthesis gas by the high-pressure process employed a catalyst system consisting of zinc oxide and chromium oxide. This catalyst, which was used at 25 – 35 MPa and 300 – 450 °C, was highly stable to the sulfur and chlorine compounds present in synthesis gas.

Production of methanol with zinc oxide –chromium oxide catalysts by the high-pressure process is no longer economical. A new generation of copper-containing catalysts with higher

activity and better selectivity is now used. The last methanol plant based on the high-pressure process closed in the mid-1980s.

3.1.3.2 Catalysts for Low-Pressure Synthesis

Well before the industrial realization of low-pressure methanol synthesis by ICI in the 1960s, copper-containing catalysts were known to be substantially more active and selective than zinc oxide – chromium oxide catalysts. Copper oxide – zinc oxide catalysts and their use in the production of methanol were described by BASF in the early 1920s. These catalysts were employed at 15 MPa and 300 °C.

Their industrial use was prevented, however, by a serious disadvantage: impurities such as hydrogen sulfide and chlorine compounds in synthesis gas rapidly deactivated the catalysts. Nevertheless, the copper-containing catalyst systems proved to be the most promising candidates for producing methanol industrially at lower temperature and pressure. A series of publications on this topic appeared between 1925 and 1955. Investigations of copper catalysts continue to this day.

A low-pressure catalyst for methanol synthesis was first used industrially in the process developed by ICI in 1966. This copper oxide – zinc oxide catalyst was thermally stabilized with alumina. It was used to convert extremely pure (i.e., largely free of sulfur and chlorine compounds, H₂S < 0.1 ppm) synthesis gas to methanol. Because this copper catalyst was extremely active, methanol synthesis could be carried out at 220 – 230 °C and 5 MPa. Premature aging due to sintering of copper was thereby avoided. The high selectivity of the new catalyst gave a methanol purity > 99.5 %. The formation of byproducts (e.g., dimethyl ether, higher alcohols, carbonyl compounds, and methane) associated with the old high-pressure catalyst, was drastically reduced or, in the case of methane, completely eliminated.

All currently used low-pressure catalysts contain copper oxide and zinc oxide with one or more stabilizing additives. Alumina, chromium oxide, or mixed oxides of zinc and aluminum have proved suitable for this purpose.

The following companies produce at the moment catalysts for the low pressure methanol synthesis:

IFP; Süd Chemie; Shell; ICI; BASF; Du Pont; United Catalysts; Haldor Topsoe

3.1.3.3 Catalyst Deactivation

Efficient catalysts for low-pressure synthesis of methanol should have a highly dispersed distribution of active centers stabilized by structural promoters. The longer a catalyst can retain these properties under industrial conditions, the more valuable it is for industrial operation: downtimes for catalyst replacement are reduced. Catalysts normally have useful lives of 2 – 5 years.

Catalyst damage and, consequently, premature deactivation may also occur during usage. The temperature conditions, hydrogen concentration of the reducing gas, and gas load must be strictly controlled. Deviations from specified reduction procedures may lead to hot spots in the pellets, resulting in sintering of the copper constituents; copper becomes mobile at 190 °C and can agglomerate from its finely divided form into fairly large crystallites. Deviations from the specified reduction conditions may permanently decrease the active BET surface area and thus irrevocably damage the catalyst.

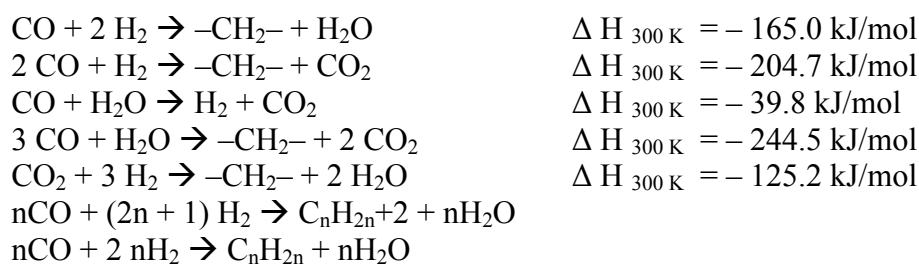
Another important point regarding the deactivation of copper catalysts is their high sensitivity to impurities in synthesis gas. Chlorine- and sulfur-containing contaminants long prevented the use of copper-containing catalyst systems in industrial methanol plants. These catalyst poisons must be removed from the feed gas prior to methanol synthesis. A certain degree of protection against deactivation by sulfur is afforded by catalysts containing zinc oxide because the sulfur is bound as zinc sulfide. After deactivation, the catalyst is still able to absorb large quantities of sulfur to protect subsequent catalyst layers against poisoning. Other synthesis gas impurities (e.g., silicon compounds, nickel carbonyls, or iron carbonyls) also cause catalyst damage.

3.1.3.4 Other Catalyst Systems

A number of modified copper – zinc oxide – alumina catalysts have been prepared by doping with boron, manganese, cerium, chromium, vanadium, magnesium, or other elements. Other catalyst systems have also been investigated. Three basic types of catalysts are mentioned in recent literature: Raney copper catalysts, copper alloys with thorium or rare-earth oxides, and supported precious-metal catalysts. Only copper alloy catalysts are reported to have a higher activity than conventional copper – zinc oxide – alumina catalysts. Until now, however, exclusively copper-containing zinc oxide – alumina catalysts have been used in industrial methanol plants. These catalysts have high activity, very good selectivity, long-term stability, and favorable production costs. They are still the most cost effective catalysts.

3.2 Fischer – Tropsch Synthesis.

Hydrogen – carbon monoxide mixtures react to hydrocarbons according to reaction equations (58) – (62), (60) is the main secondary reaction. An example for one of the general equations for alkane synthesis is (63), for olefin synthesis (64). The overall reaction balance including synthesis gas formation corresponds to an indirect coal hydrogenation.



The Fischer – Tropsch synthesis is strongly exothermic. The total heat of reaction amounts to ca. 25 % of the heat of combustion of the synthesis gas. Undesirable side reactions include methanation, the Boudouard reaction, coke deposition, oxidation of the catalyst, or carbide formation. Other competing reactions such as alcohol, aldehyde, ketone, carbonic acid, and ester formation, also occur.

Up to 1954 4000 publications and 4020 patents regarding Fischer – Tropsch catalysts had already been reported, but virtually only those based on cobalt and iron have been successful. The yield of the Fischer – Tropsch synthesis is determined by the initial hydrogen – carbon monoxide ratio and the stoichiometric requirements of the desired reaction products. The maximum yield is obtained when the aforementioned are nearly equal and thus yield 208.5 g hydrocarbon from 1 m³ (STP) of synthesis gas; technically ratios of 1.7 : 1 to 3 : 1 are used.

The Fischer – Tropsch synthesis is used on a technical scale nowadays only in South Africa at SASOL. At SASOL I, five units of the ARGE reactor of the Ruhrchemie – Lurgi (gas phase, fixed bed reactors, 2.3 – 2.5 MPa, 220 – 250 °C, 40 m³ Fe – Cu – K₂O – SiO₂ catalyst, 9 – 12 months running time) and three reactors using the Synthol circulating fluidised bed of Kellogg – SASOL (gas phase, 2.0 – 2.3 MPa, 300 – 340 °C, 30 – 140 t Fe catalyst produced by reduction of magnetite; running time 45 d) are in operation. At SASOL II (start-up 1980) and SASOL III (start-up 1982) only the more flexible Synthol process is used. The enormous gas amounts (2×1 650 000 m³ [STP]/h raw gas, 2×1 000 000 m³ [STP]/h feed gas) make the recovery of byproducts (ethylene, alcohols esters, carbonic acids, etc.) economically worthwhile.

Variations of the Fischer – Tropsch synthesis with other selective catalysts can lead to the following products:

- low-molecular-mass olefins (C₂ – C₄, mostly ethylene)
- medium- and long-chain olefins
- polyethylene

Polyethylene is obtained at high pressures (100 – 200 MPa), low temperature (100 – 120 °C), and using a ruthenium-based catalyst.

3.3 Methane Synthesis.

The catalytic hydrogenation of carbon monoxide to methane is strongly exothermic.

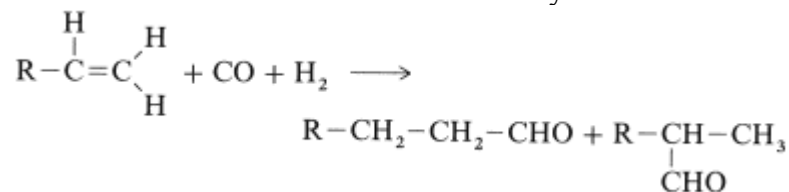


Consecutive and side reactions (shift conversion, Boudouard equilibrium, hydrogenation of carbon) make the calculation of equilibrium conditions very complex.

Classical methanation catalysts are magnesium-promoted nickel catalysts with diatomaceous earth as carrier. Catalyst developments are aimed at increasing the thermal and mechanical stability and yielding catalysts which are simultaneously suitable for the cracking of naphtha with subsequent methanation. Methanation is technically important as a catalytic purification step for the removal of carbon monoxide from gases.

3.4 Hydroformylation of Olefins.

Hydroformylation, also called oxo synthesis or Roelen reaction (O. Roelen, 1938), is a commercial-scale process for the production of aldehydes. By catalytic addition of hydrogen and carbon monoxide to an olefin an aldehyde is obtained under chain elongation



Hydroformylation is generally an exothermic, homogeneously catalyzed liquid-phase reaction of the olefin with hydrogen and carbon monoxide. As catalyst cobalt carbonyl hydride or, in some variations of the oxo-synthesis, cobalt- or rhodium-phosphin complexes are used.

Propen is the olefin mostly used. The oxoproducts are converted to alcohols, carboxylic acids, aldol-condensation products, and primary amines. About 20 commercial processes are state of the art.

3.5 Synthesis Gas as Chemical Feedstock.

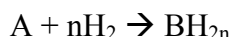
Hydrogen – carbon monoxide mixtures, hydrogen alone, and their primary product methanol are important feedstocks for the chemical industry. Nowadays, ethylene which is produced from propane, ethane, naphtha, or gas oil is the most important feedstock for the production of industrial organic chemicals in the chemical industry. Basically, it is, however, possible to obtain these compounds from synthesis gas thus changing the feedstock basis to coal.

3.6 Hydrogen in Organic Synthesis

Hydrogen is required for the production of chemicals and intermediates in organic chemistry. A large number of hydrogenations or reductions are carried out on a technical scale.

Activated and nonactivated double and triple bonds in olefins and acetylenes can be easily partially or totally hydrogenated, whereas the hydrogenation of aromatic and heterocyclic bonds requires more energetic conditions. Functional groups, such as carbonyl, nitro, nitroso, and nitrile groups, can also be hydrogenated.

The reaction conditions are dictated by equilibrium. The reactions are exothermal and run in the presence of a catalyst.



Hydrogenation catalysts are metals of groups 8 – 10 of the periodic system (see front matter of this volume), e.g., Raney nickel, as well as copper and molybdenum. In particular the noble metals (Pt, Pd), are highly-active catalysts. Homogeneous systems with molecularly dispersed catalysts in the solution, can be used for special synthesis problems (selective hydrogenation, asymmetric synthesis) but are at present of no great importance in commercial areas because of the frequently encountered difficulty to remove the catalyst from the reaction mixture.

Reaction	Product	Process features	Uses
Hydrogenation of unsaturated hydrocarbons and aromatics			
$\text{C}_6\text{H}_6 + 3 \text{H}_2 \longrightarrow \text{C}_6\text{H}_{12}$ <p>(analogous: cyclohexanol from phenol, cyclohexane carboxylic acid from benzoic acid)</p> $\text{CN}-\text{CH}_2-\text{CH}=\text{CH}-\text{CH}_2-\text{CN} + \text{H}_2 \longrightarrow \text{CN}-(\text{CH}_2)_4-\text{CN}$	cyclohexane	middle pressure hydrogenation over Ni-/Pt-Li-Al ₂ O ₃ catalyst in liquid phase (IFP, Mitsubishi) gas phase (UOP, DSM, Thoray, Houdry)	starting material for nylon production (cyclohexanone/ol, adipic acid, caprolactam), solvent
$\text{OH}-\text{CH}_2-\text{C}\equiv\text{C}-\text{CH}_2\text{OH} + 2 \text{H}_2 \longrightarrow \text{OH}-(\text{CH}_2)_4-\text{OH}$	adiponitrile	Du Pont (300 °C), liquid phase hydrogenation 25–30 MPa/70–100 °C over Raney nickel	hexanediamine
$\text{SO}_2 + \text{H}_2 \longrightarrow \text{SO}_2$	butanediol	trickle bed, 20 MPa, 180–200 °C Ni catalyst with Cu-, Cr-promoters	polyesters, polyurethane-plasticizer component, solvent
<p>Selective hydrogenation as purification step during production of ethylene, propene, and butadiene</p> <p>Further processing of oxo-products</p> $\text{R}-\overset{\text{O}}{\underset{\text{H}}{\text{C}}} + \text{H}_2 \longrightarrow \text{R}-\text{CH}_2\text{OH}$ <p>(analogous: ethylhexanol from ethylhexanal)</p> $\text{R}-\overset{\text{O}}{\underset{\text{H}}{\text{C}}} + \text{NH}_3 + \text{H}_2 \longrightarrow \text{R}-\text{CH}_2-\text{NH}_2$	sulfolane	Shell process, 11 000 t/a	aromatics extraction solvents, sour gas, scrubbing agent
	oxo-alcohols	gas phase hydrogenation at 2–0.3 MPa/115 °C, Ni catalyst	components for solvents, plasticizers, detergents
	primary amines	sump phase hydrogenation, 8 MPa/115 °C Ni catalyst	
	butanediol	hydrogenation (up to 30 MPa, 25–130 °C), Raney nickel catalyst	
	bis-(hydroxy-methyl)-cyclohexane	Mitsubishi, Kao Ind. (Japan) process	
		two step Eastman-Kodak process	
Hydrogenation of other ketones and aldehydes			
$\text{Maleic acid} \longrightarrow \text{butyrolactone} \longrightarrow \text{OH}-(\text{CH}_2)_4-\text{OH}$ $\text{CH}_3\text{OOC}-\text{C}_6\text{H}_4-\text{COOCH}_3 \xrightarrow{\text{Pd/Cu chromite}} \text{HO}-\text{CH}_2-\text{C}_6\text{H}_4-\text{CH}_2-\text{OH}$			




Reaction	Product	Process features	Uses
$\text{CH}_2=\underset{\text{H}}{\overset{\text{O}}{\text{C}}}-\text{CH} + \text{H}_2 \longrightarrow \text{CH}_2=\text{CH}-\text{CH}_2\text{OH}$	allyl alcohol	Degussa gas phase process (heterogeneous catalysis)	starting material for glycerol, glycidol
$2 \text{ Acetone} \xrightarrow{\text{Pd zeolite}} \text{mesityl oxide} \xrightarrow{i\text{-Bu}-\text{C}-\text{CH}_3}$	MIBK	hydrogenation at Pt-zeolites (modified one step processes)	(extraction) solvent
Fats, oils + H ₂ → R-CH ₂ -OH	fatty alcohols	20-40 MPa/200-400 °C, catalysts of Adkins type	
R-O-CH ₂ CH ₂ -OH + HCHO + H ₂ → R-O-CH ₂ CH ₂ -OCH ₃	polyethylene-glycol ethers	new Hoechst process	sour gas scrubbing agent (Selexol)
Hydrogenation of N-compounds			
	aniline	fixed bed hydrogenation over NiS/CuS, 300-475 °C (Bayer, Allied, Lonza), fluidized bed hydrogenation, 5 MPa/100 °C over Cu catalyst (BASF, Cyanamid)	starting material for dyes, pharmaceuticals, isocyanate polymers, solvents
	diaminotoluenes	analogous to nitrobenzene reduction	
Nitriles + H ₂ → primary amines	hexanediamine	5-15 MPa/60-130 °C over Raney Ni or Raney Co	starting material for fibers, sour gas scrubbing agent
Miscellaneous reactions			
	benzene	3-5 MPa/500-650 °C over Cr ₂ O ₃ -Mo ₂ O ₃ -CoO catalyst (Houdry, UOP, Shell, BASF) 10-25 MPa/400-500 °C over Pt-Al ₂ O ₃ -SiO ₂	50% of toluene production further processed to benzene
Xylene isomerization under H ₂ partial pressure	o-/p-xylene	catalysts	terephthalic acid, phthalic acid

Table (40) gives an overview of commercially used hydrogenation reactions.

High-purity hydrogen is necessary for the partial or total hydrogenation of fats and oils (for the production of edible fats or for technical purposes). In fat hydrogenation the polyene, triene, and diene fatty acids in their glyceride form are selectively hydrogenated to the corresponding monoene acids.

The industrial production of sugar alcohols, such as sorbitol, xylitol or mannitol from the corresponding sugars is carried out by catalytic hydrogenation. Batch suspension processes using Raney nickel catalysts are mainly employed under reaction conditions of 120 – 150 °C and 3 – 7 MPa.

3.7 Hydrogen in Inorganic Synthesis

The catalytic hydrogenation of anthraquinone and its derivatives followed by their auto-oxidation to yield hydrogen peroxide is the basis of the commercially important process for hydrogen peroxide production. Further important reactions in inorganic chemistry are the production of hydrochloric acid from hydrogen and chlorine and the hydroxylamine synthesis