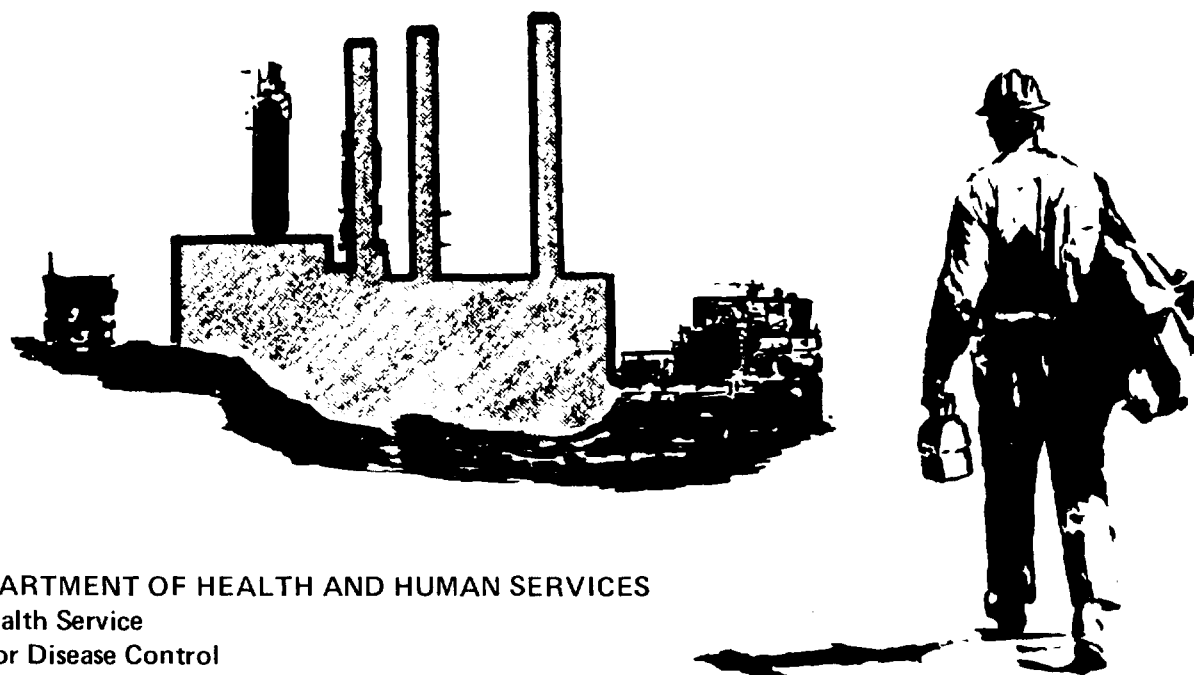


NIOSH

OCCUPATIONAL HAZARD ASSESSMENT

COAL LIQUEFACTION, Volume II - Assessment



U.S. DEPARTMENT OF HEALTH AND HUMAN SERVICES
Public Health Service
Centers for Disease Control
National Institute for Occupational Safety and Health

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Volume II - Assessment

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PREFACE

The National Institute for Occupational Safety and Health (NIOSH) believes that coal liquefaction technology presents potential hazards to workers because of similarities with other coal-related processes that have shown high cancer risks. This occupational hazard assessment critically reviews the scientific and technical information available and discusses the occupational safety and health issues of coal liquefaction pilot plant operations. By addressing the hazards while the technology is in the developmental stage, the risk of potential adverse health effects can be substantially reduced in both experimental and commercial plants.

This occupational hazard assessment is intended for use by organized labor, industry, trade associations, government agencies, and scientific and technical investigators, as well as the interested public. The information and recommendations presented in this assessment should facilitate the development of specific procedures for hazard control in individual workplaces by those persons immediately responsible for health and safety. NIOSH will periodically update and evaluate new data and information as they become available and, at the appropriate time, will consider proposing recommendations for a standard to protect workers in commercial coal liquefaction facilities.



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I. EXECUTIVE SUMMARY

Coal liquefaction is one of the technologies now being developed in the United States to offset increasing energy demands. For commercial application of this technology, engineering design progresses through the stages of bench-scale units, process development units, pilot plants, and demonstration plants to commercial plants. Today, several coal liquefaction pilot plants, with varying capacities, are operating in the United States. Other pilot and demonstration plants that will have larger capacities are being constructed or designed [1].

This document reviews and evaluates potential occupational hazards to workers in coal liquefaction pilot plants. By addressing the hazard while direct coal liquefaction technology is still being developed, the risk of potentially adverse health effects can be substantially reduced. In this document recommendations are made for reducing the risks of these adverse health effects. Failure to take adequate precautionary measures may result in needless risks to worker health and safety.

Coal liquefaction is the conversion of coal to liquid hydrocarbon products. The major products of most coal liquefaction processes are condensed aromatic liquids; however, some gases and solids are also produced, depending on the type of coal, the process, and the operating conditions used [2].

Specific coal liquefaction processes differ in the methods and operating conditions used to break physical and chemical bonds, in the sources of hydrogen used to stabilize radical fragments, and in the physical and chemical characteristics of product liquids, gases, and solids. There are four categories of coal conversion processes: (1) pyrolysis, (2) solvent extraction, (3) direct hydrogenation, and (4) indirect liquefaction [3]. This assessment is concerned with direct liquefaction, ie, processes 1-3. Although equipment within a coal liquefaction plant varies according to the processes employed, there are many similarities. Some operations common to the plants include coal handling and preparation, liquefaction, physical separation, upgrading, product storage, and waste management.

Coal liquefaction materials contain potentially hazardous biologically active substances. Skin cancers were reported among workers in one coal hydrogenation pilot plant that is no longer operating [4]. Evidence from animal experiments indicates that local skin carcinomas may result when some coal liquefaction products remain on the skin for long periods of time [5-9]. Similarities exist between the toxic potential of coal liquefaction products and that of other materials derived from coal, such as coal tars, coal tar pitch, creosote, and coke oven emissions, which have been associated with a high cancer risk. Some compounds, such as benzo(a)pyrene, methyl chrysenes, aromatic amines, and certain other polycyclic aromatic hydrocarbons, that are known human carcinogens when they occur individually were found in pilot plant

products and process streams [10-13]. The carcinogenic potential of these compounds when they occur in mixtures is unknown.

In addition to the carcinogenic potential of constituent chemicals in various coal liquefaction process streams, other long-term effects on nearly all major organ systems of the body have been attributed to them. Many of the aromatics and phenols irritate the skin or cause dermatitis. Silica dust and other components of the mineral residue may affect the respiratory system. Benzene, inorganic lead, and nitrogen oxides may affect the blood. Creosotes and coal tars affect the liver and kidneys, and toluene, xylene, hydrogen sulfide, and inorganic lead may affect the central nervous system (CNS). Evidence from recent animal studies [14,15] also indicates that coal liquefaction materials may have adverse effects on reproduction. The potential also exists for worker exposure to hazards that are an immediate threat to life, such as hydrogen sulfide, carbon monoxide, and fire and explosion.

The recommendations made in this document for worker protection include a combination of engineering controls, work practices, personal protective equipment, and medical surveillance. Additional recommendations for training, emergency procedures, and recordkeeping are made to support the engineering control and work practice recommendations. Although insufficient data are available at this time to support recommending environmental exposure limits for all materials found in coal liquefaction processes, some information is available from similar industries [16-18].

The primary objectives of engineering controls are to minimize the potential for worker exposure to hazardous materials and to reduce exposure levels. Design considerations should ensure the integrity of process containment; limit the need for worker exposure; provide for maximum equipment reliability; minimize the effects of erosion, corrosion, instrument failure, and seal and valve failure; and provide for equipment separation, redundancy, and fail-safe design.

The major objective of recommended work practices is to provide additional protection to the worker when engineering controls are not adequate or feasible. Most coal liquefaction pilot plants have written policies and procedures for various work practices, including breaking into pipelines, lockout of electrical equipment, tag-out of valves, fire and rescue brigades, safe work permits, vessel entry permits, wearing safety glasses and hardhats, housekeeping, safe storage of process materials, decontamination of equipment requiring maintenance, and other operational safety practices [1].

Personal protective equipment such as respirators and protective clothing may be necessary to prevent worker exposure to coal-derived materials. However, they should be used only when other methods of control are inadequate.

Because workers in coal liquefaction plants may be exposed to a wide variety of chemicals that can produce adverse health effects, medical surveillance is necessary to evaluate the ability of workers to perform their work

and to monitor them for any changes or adverse effects. Particular attention should be paid to the skin, oral cavity, respiratory system, and CNS. NIOSH recommends that a surveillance program be instituted that includes preplacement, periodic, and termination physical examinations as well as preplacement and interim medical histories.

Sampling and analysis for air contaminants provide a way to assess the performance of engineering controls. Industrial hygiene monitoring can be used to determine employee exposure to chemical and physical hazards. The combination of data from exposure records, work histories, and medical histories provides a way to evaluate the effectiveness of engineering controls and work practices, and to identify causative agents for effects that may be revealed during medical monitoring. Thus, it is important that medical records and pertinent supporting documents be established and maintained for all workers and that copies of any applicable environmental exposure records be included.

At the beginning of employment, all workers should be informed of the occupational exposure hazards associated with coal liquefaction plants. As part of a continuing education program, training should be repeated periodically to ensure that all employees have current knowledge of job hazards, signs and symptoms of overexposure, proper maintenance and emergency procedures, proper use of protective clothing and equipment, and the advantages of good personal hygiene.

The data used in this occupational hazard assessment were obtained and evaluated through literature surveys and from visits to coal liquefaction pilot plants or related facilities. Data from industries in which workers have been exposed to materials similar to those found in coal liquefaction plants were also considered. Acronyms used in the document are listed in Chapter XX.

II. COAL LIQUEFACTION PROCESS TECHNOLOGY

Coal Conversion

(a) Background

Coal can be converted into synthetic fuels by coal gasification or liquefaction processes, mainly yielding a gas or liquid, respectively. However, both gaseous and liquid products and byproducts can be obtained from most gasification and liquefaction processes [3]. In addition, similar equipment, eg, gas purification systems and coal handling equipment, can be found in both types of processes. Where these similarities exist, NIOSH's previous recommendations in the criteria document on coal gasification plants are applicable [16].

Examples of equipment generally found in coal liquefaction plants, but not in gasification plants, include dissolvers, catalytic hydrogenation reactors, solid-liquid separation units, and solvent recovery units. Unlike coal gasification plants, coal liquefaction plants process coal-oil slurries at high pressures and temperatures. This operating environment presents the potential for erosion, corrosion, and seal failures, resulting in the release of flammable hydrocarbon liquids and/or other hazardous materials. Another problem in liquefaction is plugging associated with solidification of the coal solution when its temperature drops to less than the pour point of the mixture.

Coal gasification entails treatment of coal in a reducing atmosphere with air or oxygen, steam, carbon monoxide, hydrogen, or mixtures of these gases to yield a combustible material [16]. The primary product from gasification is a mixture of hydrogen, water, carbon monoxide, carbon dioxide, methane, inerts (eg, nitrogen), and minor amounts of hydrocarbons and other impurities [16]. Hydrogen and carbon monoxide are then catalytically treated to produce pipeline-quality gas and light oils. In a gasification process, liquid byproducts may be recycled to the reactor while gaseous products are cleaned, upgraded, and stored or shipped [3].

Coal liquefaction is the process that converts coal to liquid hydrocarbon products. Some gases and solids are also produced, depending on the type of coal, the process, and the operating conditions used. In general, the changes that occur in the liquefaction of coal include breaking weak van der Waal's forces and hydrogen bonds between layers in the coal structure, rupturing both aromatic-aromatic and aromatic-aliphatic chemical bonds, and stabilizing free radical fragments [2]. Although there are exceptions, the major products of most coal liquefaction processes are condensed aromatic liquids [2].

Although similarities in equipment exist, the hazards associated with each type of process were assessed independently [16]. Two important differences

in health and safety hazards between the two processes are (1) the chemical composition of products and process streams, which may affect overall health risks; and (2) equipment configuration, which may affect the potential for release of process materials.

(b) Coal Liquefaction Processes

Specific coal liquefaction processes differ in the methods and operating conditions used to break physical and chemical bonds, in the sources of hydrogen used to stabilize radical fragments, and in the physical and chemical characteristics of product liquids, gases, and solids. Significant features of the four major categories of coal conversion processes are discussed below.

(1) Pyrolysis

Pyrolysis involves heating coal to a temperature between 400 and 550°C in the absence of air or oxygen, resulting in disruption of physical and chemical bonds, generation of radicals, and abstraction of hydrogen atoms by radicals for coal hydrogen-donors. During this process, some small radicals combine to form hydrogen-enriched volatile hydrocarbon components. Loss of donor-hydrogen from larger fragments produces char. Pyrolysis products include heavy oil, fuel oil, char, and hydrocarbon gases. Temperatures greater than 550°C promote cracking and high gas yields.

Pyrolysis in the presence of hydrogen, at or above atmospheric pressure, is known as hydrocarbonization. Generally, hydrocarbonization products are similar to those obtained by simple pyrolysis, but are somewhat lower in char yield.

(2) Solvent Extraction

Solvent extraction processes are generally performed at high temperatures and pressures in the presence of hydrogen and a process-derived solvent that may or may not be hydrogenated. The solvent-refined coal (SRC) process produces either liquid or solid low-ash and low-sulfur fuels, depending on the amount of hydrogen introduced. The liquid is used as a boiler fuel. The Exxon donor-solvent (EDS) process produces gases and liquid fuels from a wide variety of coals.

(3) Direct Hydrogenation

Direct hydrogenation is a process in which a coal slurry is hydrogenated in contact with a catalyst under high temperatures and pressures. Process products are boiler fuels, synthetic crude, fuel oil, and some gases, depending on process conditions.

(4) Indirect Liquefaction

In indirect liquefaction, carbon monoxide and hydrogen produced by gasifying coal with steam and oxygen can be catalytically converted into

liquid fuels. Another indirect catalytic liquefaction process produces methanol, which can be converted to gasoline.

(c) Process Development

Significant technical advances in coal liquefaction were made in Germany between 1915 and 1944 [19]. Germany developed and improved the Bergius coal liquefaction process, which consisted of hydrogenating finely ground coal by amalgamation with tar oils. Product oil was fractionated by distillation, and the heavy fraction provided the tar oil used to hydrogenate the finely ground coal. The light fraction was upgraded by using hydrogen-enriched steam to produce a liquid rich in aromatics [19]. During World War II, the Germans constructed 11 hydrogenation plants in addition to 7 existing plants. In 1944, the total output capacity of these 18 coal hydrogenation plants was 4 million metric tons (4 Tg) of oil a year. These plants supplied almost all of the fuel necessary for German aviation in 1944 [3].

Another coal liquefaction process was developed in the 1920's by Fischer and Tropsch [19]. This Fischer-Tropsch process uses synthesis gas, formed by passing steam over red-hot coke, to produce liquid hydrocarbons in a catalytic reaction. Currently, this process is being used on a commercial scale at the South African Coal, Oil, and Gas Corporation, Ltd (SASOL) plant in South Africa (SASOL I) [20,21]. In addition, South Africa is currently operating a second plant (SASOL II), and a third plant (SASOL III) is scheduled to be operating by 1984. The production capacity of the SASOL II plant is estimated to be 2.1 million metric tons (2.1 Tg) of marketable products per year [22]. Of this figure, SASOL estimates that motor fuels production will be 1.5 million metric tons (1.5 Tg) per year [3,22]. Currently, SASOL I total output is approximately 0.25 million metric tons (0.25 Tg) of petrochemicals per year, which includes 0.168 million metric tons (0.168 Tg) of gasoline [3].

Coal liquefaction experience in the United States [20] includes (1) synthetic oil research conducted at the Pittsburgh Energy Technology Center (PETC) (formerly Pittsburgh Energy Research Center) since the early 1950's, (2) a coal liquefaction demonstration plant using the Bergius process, which operated in the 1950's in Louisiana, Missouri, (3) a hydrogenation pilot plant operated by Union Carbide from 1952 to 1959 at Institute, West Virginia, (4) char-oil-energy development (COED) process development begun in 1962 by FMC Corporation, (5) Consolidation Coal Company development of Consol synthetic fuel (CSF) process begun in 1963, (6) Hydrocarbon Research, Inc, H-coal process begun in 1964, (7) SRC research initiated by the Office of Coal Research (OCR) in 1962, and (8) donor-solvent research started by Exxon in 1966 [3]. Congressional authorization bills for FY 76, 77, and 78 have provided approximately \$100 million annually in Federal funding for coal liquefaction research and development [23]. More than \$200 million annually has been authorized for FY 79, 80, and 81 [24,25].

Coal liquefaction operations in the United States have been limited to bench-scale units, process development units, and pilot plants capable of handling up to 600 tons of coal per day (545 Mg/d) [1]. However, a commercial plant that could process approximately 30,000 tons (27,000 Mg) of coal per day is envisioned for the late 1980's [26]. In addition to being larger than pilot plants, commercial plants will be designed and operated differently [1,26].

Pilot plants are used to assess the feasibility of process technology and operability of equipment such as pumps, seals, and solid-liquid separation units. They are also used to optimize operating conditions. To obtain the necessary data, production runs are of shorter duration than those in commercial plants, which are designed to operate continuously. Commercial plants are designed to economically produce marketable products. For this reason, commercial plants may recycle treated wastewater products and/or use char, mineral residue slurry, and sulfur byproducts [1,26,27], whereas most pilot plants currently do not. Equipment may differ based on the development of new technology, eg, new solid-liquid solvent de-ashing separation units currently being tested [1,28,29]. Some equipment used in pilot plants, eg, a rotating liquid/solid extractor or a mineral residue dryer, may not be feasible or necessary in commercial plants [1].

Although commercial plant design and equipment may differ from that of pilot plants, the engineering design considerations that may affect the potential for worker exposure may be similar. Both commercial and pilot plants will operate in an environment of high temperature and pressure, and in most cases, a coal slurry will also be used under these conditions [26,30]. The types of exposure resulting from leaks, spills, maintenance, handling, and accidents may be qualitatively similar for both commercial and pilot plants although frequency and duration of exposure may vary [1]. Specific control technology used to minimize worker exposure may differ in both types of plants. For example, due to the continuous operating mode of a commercial plant, a closed system may be used to handle solid wastes in order to minimize inhalation hazards. This system may not be economical for a pilot plant with batch operations, because portable local exhaust ventilation could be provided when needed [1]. Both of these systems are designed to minimize worker exposure to hazardous materials.

Description of General Technology

The Pott-Broche and Bergius processes were forerunners of two liquefaction processes under development in the United States [2,31]. These developmental processes, discussed at the beginning of the chapter, are categorized as solvent extraction and hydrogenation. Pyrolysis and indirect liquefaction are two additional processes currently used. Figure XVIII-1 illustrates the possible coal liquefaction routes. There is no clear distinction between the solvent extraction and hydrogenation categories [32]. Solvent extraction is generally grouped with noncatalytic hydrogenation [2,20,30,32], while in

other studies catalytic and noncatalytic hydrogenation appear under one category, ie, hydrogenation [19,31,33,34]. The latter categorization is used in this assessment.

Coal liquefaction processes using solvent extraction, hydrogenation, pyrolysis/hydrocarbonization, and indirect liquefaction are discussed in Appendix I. Specific processes discussed are the CSF, SRC, H-coal, COED, and Fischer-Tropsch processes, respectively. Appendix II summarizes the major coal liquefaction systems under development in the United States. This assessment does not address the necessary controls and work practices for indirect liquefaction processes, eg, the SASOL technology, since they were previously evaluated by NIOSH [16]. Commercial plants using a process similar to the SASOL technology should follow the recommendations contained in the NIOSH coal gasification criteria document [16].

Although systems and components vary according to the process employed, there are similarities between most coal liquefaction plants. Systems common to coal liquefaction plants include coal handling and preparation, liquefaction, physical separation, upgrading, product storage, and waste management. Appendix III shows the applicability of these major systems to the various coal liquefaction processes summarized in Appendix II. Appendix IV lists the major equipment used in coal liquefaction and a description of its function.

Figure XVIII-2 is a schematic of the general systems used in coal liquefaction. Not all of the unit operations/unit processes shown are applicable to each coal liquefaction process.

(a) Coal Handling and Preparation

The purpose of the coal handling and preparation system is to receive run-of-mine (ROM) coal and prepare it for injection into the liquefaction system. This front-end process is basically the same in all liquefaction plants and produces pulverized coal and coal slurry. Dusts, coal fines, and solvents also may be present. ROM coal is received by rail or truck and is dumped into receiving hoppers. The coal is crushed and transferred to storage bins. When needed, the coal is retrieved from storage, pulverized and dried, and transferred to a blend tank where it is mixed with process solvent to form a coal slurry. At this point, the coal is pumped into the liquefaction system. The slurry blending step is essential for solvent extraction, and catalytic and noncatalytic hydrogenation processes. However, this step is omitted in pyrolysis processes, in which pulverized coal is fed directly into the reactor usually by means of lockhoppers.

(b) Liquefaction

The function of a liquefaction system is to transform coal into a liquid. Solvent extraction and catalytic and noncatalytic hydrogenation are three-phase systems that involve the use of significant quantities of hydrogen [2,35]. Pyrolysis is a two-phase, ie, solid-gas, system. If hydrogen is

added during pyrolysis, the process is called hydrocarbonization. Temperatures in these systems range from 700 to 1,500°F (371 to 820°C); pyrolysis reactors generally operate in the upper range [2,35]. Materials found within the liquefaction system include hydrogen, recycled and makeup solvent, gases (hydrogen sulfide, carbon monoxide, methane), solids (unreacted coal, char, ash, catalyst), coal slurries, and organic liquid fractions of the product.

(c) Separation

The product stream from liquefaction contains a mixture of gases, vapors, liquids, and solids and is typically fed to a gas-liquid separator such as a flash drum. Here the pressure on the product stream is reduced, allowing the lower boiling chemicals to vaporize and gases to separate from the liquid. These vapors and gases are separated in a condensate system that removes the higher boiling components of the gas stream. The solids are separated from the liquids by such processes as filtration, centrifugation, distillation, or solvent de-ashing. Materials found in the separation systems include solvents, gases (carbon dioxide, hydrogen sulfide, hydrogen, methane), water, light oils, heavy oils, and solids (mineral residue, unreacted coal).

(d) Upgrading and Gas Purification

The upgrading and gas purification system refines and improves the gases and liquids obtained from the separation system. A gas desulfurization unit removes the sulfur from the gases. The hydrocarbon gases may be further upgraded by methanation to produce pipeline-quality gas or are sent to a hydrogen-methane separation unit where the resulting hydrogen could be used for hydrogenation [3]. The liquid stream may be upgraded by fractionation, distillation, hydrogenation, or a combination of these, resulting in products such as synthetic oils and solvent-refined coal.

(e) Product Storage

Gas products from the liquefaction plant can be stored onsite in tanks or can be piped directly offsite. If piped offsite, there could be reserve storage to allow for peak demands for the product. The liquid products can be stored in tanks, tank cars, or trucks or, as in the case of solvent-refined coal, can be solidified by using a prilling tower or a cooling belt. Depending on its biological and chemical properties, the solid product could be stored in open or closed storage piles.

(f) Waste Management

The waste management system includes gas scrubbers, settling ponds, and wastewater treatment facilities. Its function is to reduce pollutants in the waste streams in accordance with discharge regulations established by Federal, State, and local environmental protection agencies. Typical plant-produced

wastes that must be treated and disposed of include solids such as coal particulate, ash, slag, mineral matter, sludges, char, and spent catalyst; wastewater containing suspended particles, phenols, tars, ammonia, chlorides, and oils; and gases such as carbon monoxide, hydrogen sulfide, and hydrocarbon vapors [31]. Waste treatment facilities are also designed to collect and treat process materials released by spills.