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Carbon Disulfide Emission Control Options

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Carbon Disulfide Emission Control Options

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Carbon Disulfide Emission Control Options

by

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PREFACE

This project was funded by EPA's Control Technology Center (CTC) and prepared by TRC Environmental Corporation (TRC).

The CTC was established by EPA's Office of Research and Development (ORD) and Office of Air Quality Planning and Standards (OAQPS) to provide technical assistance to State and local air pollution control agencies. Several levels of assistance can be accessed through the CTC. First, a CTC Hotline has been established to provide telephone assistance on matters relating to air pollution control technologies. Second, more in-depth engineering assistance can be provided when appropriate. Third, the CTC can provide technical guidance through designing technical guidance documents, developing personal computer software, and presenting workshops on control technology matters. The CTC is also the focal point of the Federal Small Business Assistance Program, and maintains the Reasonably Available Control Technology/Best Available Control Technology/Lowest Achievable Emission Rate (RACT/BACT/LAER) Clearinghouse and the Global Greenhouse Gases Technology Transfer Center. Information concerning all CTC products and services can be accessed through the CTC Bulletin Board System (BBS), which is part of the OAQPS Technology Transfer Network (TTN) bulletin board system.

The technical guidance projects, such as this one, focus on topics of national or regional interest and are identified through contact with State and local agencies or private organizations. In this case, the CTC Hotline was accessed by numerous agencies seeking information on controlling carbon disulfide emissions. The interest in this topic warranted development of a technical document on carbon disulfide emissions control options. This document briefly discusses various options available to public and private industry for controlling carbon disulfide emissions, and serves as a reference source for those seeking further information.

TABLE OF CONTENTS

Section		Page
DISCLAIMER		ii
ACKNOWLEDGEMENTS		iii
PREFACE		iv
LIST OF FIGURES		vii
1.0	INTRODUCTION	1
1.1	Purpose of Report	1
1.2	Project Methodology	1
2.0	CARBON DISULFIDE: BACKGROUND INFORMATION	3
2.1	Physical Properties	3
2.2	Chemical Properties	4
2.3	Production and End-Uses	4
2.4	Health Effects Summary	5
3.0	EMISSION SOURCES FOR CARBON DISULFIDE	7
3.1	Viscose Process	7
3.2	Carbon Tetrachloride Production	9
3.3	Other Processes	10
4.0	CONTROL TECHNIQUES	12
4.1	Absorption	12
4.1.1	Method 1: Oil Absorption	12
4.1.2	Method 2: International Patent No. WO 86/02283	13
4.1.3	Method 3: Diglycolamine Agent	16
4.1.4	Conclusions	16
4.2	Adsorption	17
4.2.1	Method 1: Activated Carbon	17
4.2.2	Method 2: Synthesized Sorbent: European Patent EP 189606	20
4.2.3	Conclusions	21
4.3	Ventilation and Condensation Recovery System	21
4.4	Absorption/Oxidation: U.S. Patent 4,049,775	24
4.5	Regenerative Thermal Oxidation	26
4.6	Catalytic Conversion: U.S. Patent 4,374,819	27

TABLE OF CONTENTS (continued)

Section | Page

4.7 Control with a Bioreactor 31
4.8 Wet Scrubbing 31
4.9 Current Research 33

REFERENCES 36

LIST OF FIGURES

Number		Page
1.	Viscose Process	8
2.	Oil Absorption System	11
3.	Carbon Tetrachloride Production	14
4.	Adsorption Recovery System	19
5.	Ventilation and Recovery System	22
6.	BASF Carbon Disulfide Recovery System	23
7.	Wet Method of Carbon Disulfide Control	25
8.	Thermal Oxidation System	28
9.	Catalytic Conversion	30
10.	Control With a Bioreactor	32
11.	Wet Scrubbing	34

CARBON DISULFIDE CONTROL OPTIONS

1.0 INTRODUCTION

1.1 Purpose of Report

The EPA Control Technology Center (CTC) has received a number of inquiries regarding control options for carbon disulfide emissions. This report presents a description of methods or techniques to control carbon disulfide emissions from a variety of source categories (Section 4.0). The control techniques presented represent potential control options, as many of them have not yet been proven in an industrial environment. The report also contains background information on carbon disulfide (Section 2.0) and a discussion of carbon disulfide emission source categories (Section 3.0).

1.2 Project Methodology

The project was initiated by a comprehensive literature search involving several computer databases (i.e., Chemical Abstracts, PTS-Prompt, Chemical Economics Handbook, Kirk-Othmer). The computer search provided information primarily on foreign applications by locating patents related to control of carbon disulfide emissions. The reader should be aware that these patents may represent carbon disulfide emissions control options that are still in research or experimental stages. The computer search was also able to provide some information on domestic applications; however, the bulk of the information for domestic applications came from discussions with vendors and plant operators.

Vendors were able to supply substantial information on equipment and solvents that they felt may be used as control options. Industry, however, was very conservative with information about their process conditions and particular control configurations. It should be noted that any information obtained from plant operators may not be complete to the extent of supplying

technical process descriptions/diagrams. The Chemical Manufacturer's Association (CMA) provided significant information for this document.

2.0 CARBON DISULFIDE: BACKGROUND INFORMATION

The following four subsections outline the physical/chemical properties of carbon disulfide, current production information and trends, major end-uses of carbon disulfide, and health effects of carbon disulfide exposure.

2.1 Physical Properties

Carbon disulfide is a clear and colorless liquid with a mild ethereal odor; however, minor impurities impart a disagreeable sulfurous odor. It is slightly soluble in water and an excellent solvent for many organic compounds. The physical properties are listed below (1,2,3).

PROPERTY		VALUE	
melting pt, K		161.11 (-169.7°F)	
latent heat of fusion kJ/kg		57.7 (5148 cal/lb)	
boiling pt at 101kPa,°C		46.25 (114.8°F)	
flash point at 101kPa,°C		-30 (-22°F)	
ignition temp in air, °C			
10-s lag time		80-120 (176-248°F)	
0.5-s lag time		156 (312.8°F)	
critical temp, °C		273 (523.4°F)	
critical density, kg/m^3		378 (28.7 lb/ft^3)	
critical pressure, k/Pa		7700 (76 atm)	
Liquid at temp, °C	**0 (32°F)**	**20 (68°F)**	**46.25 (114.8°F)**
density, kg/m^3	1293 (98.1 lb/ft^3)	1263 (95.8 lb/ft^3)	1224 (92.9 lb/ft^3)
specific heat, J(kgxk)	984	1005	1030
latent heat of vap kJ/kg	377 (33634 cal/lb)	368 (32832 cal/lb)	355 (31671 cal/lb)
surface tension, mN/m^3	35.3 (35.3 dynes/cm)	32.3 (32.3 dynes/cm)	28.5 (28.5 dynes/cm)
thermal conductivity, W/mxK		0.161	
viscosity, mPaxs	0.429	0.367	0.305
refractive index	1.6436	1.6276	
soly in H_2O, g/kg sol'n	2.42	2.10	0.48
vapor pressure kPa	16.97 (0.17 atm)	39.66 (0.39 atm)	101.33 (1.00 atm)
Gas at temp, °C	**46.25 (114.8°F)**	**200 (392°F)**	**400 (752°F)**
density, kg/m^3	2.97 (0.19 lb/ft3)	1.96 (0.12 lb/ft^3)	1.37 (0.09 lb/ft^3)
specific heat, J(kgxK)	611	679	730
viscosity, mPaxs	0.0111	0.0164	0.0234
thermal conductivity, W/mxK	0.0073		
Thermochemical data at 298 K (77°F)			
heat capacity, Cp, J(molxK)		45.48	
entropy, S, J(molxK)		237.8	
heat of formation, Hf, kJ/mol		117.1 (27,988 cal/mol)	
free energy of formation, kJ/mol		66.9 (15,989 cal/mol)	

Carbon disulfide is soluble in most organic solvents and completely miscible with many liquid hydrocarbons, chlorinated hydrocarbons, and alcohols.

2.2 Chemical Properties

Carbon disulfide is highly flammable and is easily ignited by sparks or hot surfaces. Greater availability of oxygen or lower pressure may cause a considerable lowering of the flash point temperature. The flammability or explosive range of carbon disulfide with air is very wide and depends upon the conditions. Concentrations of 4 to 8 percent carbon disulfide in air explode with maximum violence. The upper explosive limit (UEL) and lower explosive limit (LEL) for carbon disulfide in air are 50 percent and 1 percent, respectively (4). In practice, for safety reasons control equipment must be designed to operate above the UEL or below the LEL. The thermal energy released in carbon disulfide explosions is low compared to that from other flammable substances. The maximum absolute pressure developed is reported as 739 kPa (7.30 atm).

2.3 Production and End-Uses

In 1989, carbon disulfide was primarily used as a chemical intermediate, and production was estimated to be 340 million pounds. At one time, carbon disulfide was used mainly as a solvent. However, most carbon disulfide is currently used in the synthesis of other chemical products. Approximately 40 percent of U.S. production of carbon disulfide is used in the viscose process of regenerating cotton and wood cellulose to make rayon and cellophane. Carbon tetrachloride production also accounts for over 40 percent of U.S. consumption of carbon disulfide. This usage level of carbon disulfide is expected to decline through the 1990s, because the demand for carbon tetrachloride is expected to decline (its predominant end use is in the production of fluorocarbon gases). Other products made from carbon disulfide include rubber

accelerators for vulcanization, mercaptans added to fuel gases to impart detectable odors, certain synthetic polymers, oils, fats and waxes. It is also used as a solvent for paraffins and waxes in oil wells because it is relatively non-corrosive.

The end use pattern for carbon disulfide in 1989 is listed below (2).

Derivative	Percentage
Carbon tetrachloride	43
Rayon	35
Cellophane	4
Rubber chemicals	9
Misc.	9

2.4 Health Effects Summary

No data exist that classify carbon disulfide as an animal or human carcinogen. However, at very high levels (10,000 ppm carbon disulfide in air), carbon disulfide is highly toxic to the central nervous system, and death can occur due to respiratory failure. Carbon disulfide can cause neurotoxic effects through inhalation and dermal contact (1,5). Carbon disulfide can also be highly toxic when ingested in large doses (2).

Various local effects and symptoms may occur from exposure to carbon disulfide. In sufficient quantities, carbon disulfide vapor is severely irritating to eyes, skin, and mucous membranes. Permanent visual impairment and retinal cell degeneration occurred in monkeys that breathed 256 ppm for 6 hours a day, 5 days a week for 5-13 months (1). Contact with liquid carbon disulfide may cause blistering with second and third degree burns. Skin sensitization may occur. Skin absorption may result in localized deterioration of peripheral nerves, most often noted in the hands. Respiratory irritation may result in bronchitis and emphysema (6).

Subjective psychological and behavioral disorders have been observed following repeated carbon disulfide exposure. Acute exposures may result in extreme irritability, uncontrollable

anger, suicidal tendencies, and a toxic manic depression psychosis. Chronic exposures have resulted in insomnia, nightmares, impaired memory, and impotency. Less severe changes include headache, dizziness, and diminished neutral and motor ability, with staggering gait and loss of coordination (6).

Atherosclerosis and coronary heart disease have been significantly linked to carbon disulfide exposure. A significant increase in coronary heart disease mortality has been observed in carbon disulfide workers. The Federal Occupational Safety and Health Administration (OSHA) has determined that a 4 ppm 8-hour time-weighted average (TWA) limit and a 12-ppm STEL (Short-Term Exposure Limit) are necessary to reduce the risk of cardiovascular disease among carbon disulfide-exposed workers. Studies also reveal higher frequency of angina pectoris and hypertension (6,7).

Other specific effects of chronic carbon disulfide exposure include chronic gastritis with the possible development of gastric and duodenal ulcers; impairment of endocrine activity, specifically adrenal and testicular; abnormal erythrocytic development with hypochromic anemia; and possible liver dysfunction with abnormal serum cholesterol. Chronic menstrual disorder may occur in women. These effects usually occur as a result of chronic exposure of carbon disulfide and are subordinate to the other symptoms (6).

3.0 EMISSION SOURCES FOR CARBON DISULFIDE

This section presents information on the main source categories that emit carbon disulfide (i.e., rayon production and carbon tetrachloride production)(2,3,8,9). Included in the discussion of each source category is a technical description of the emitting process and, where available, a mention of the specific emission points.

3.1 Viscose Process

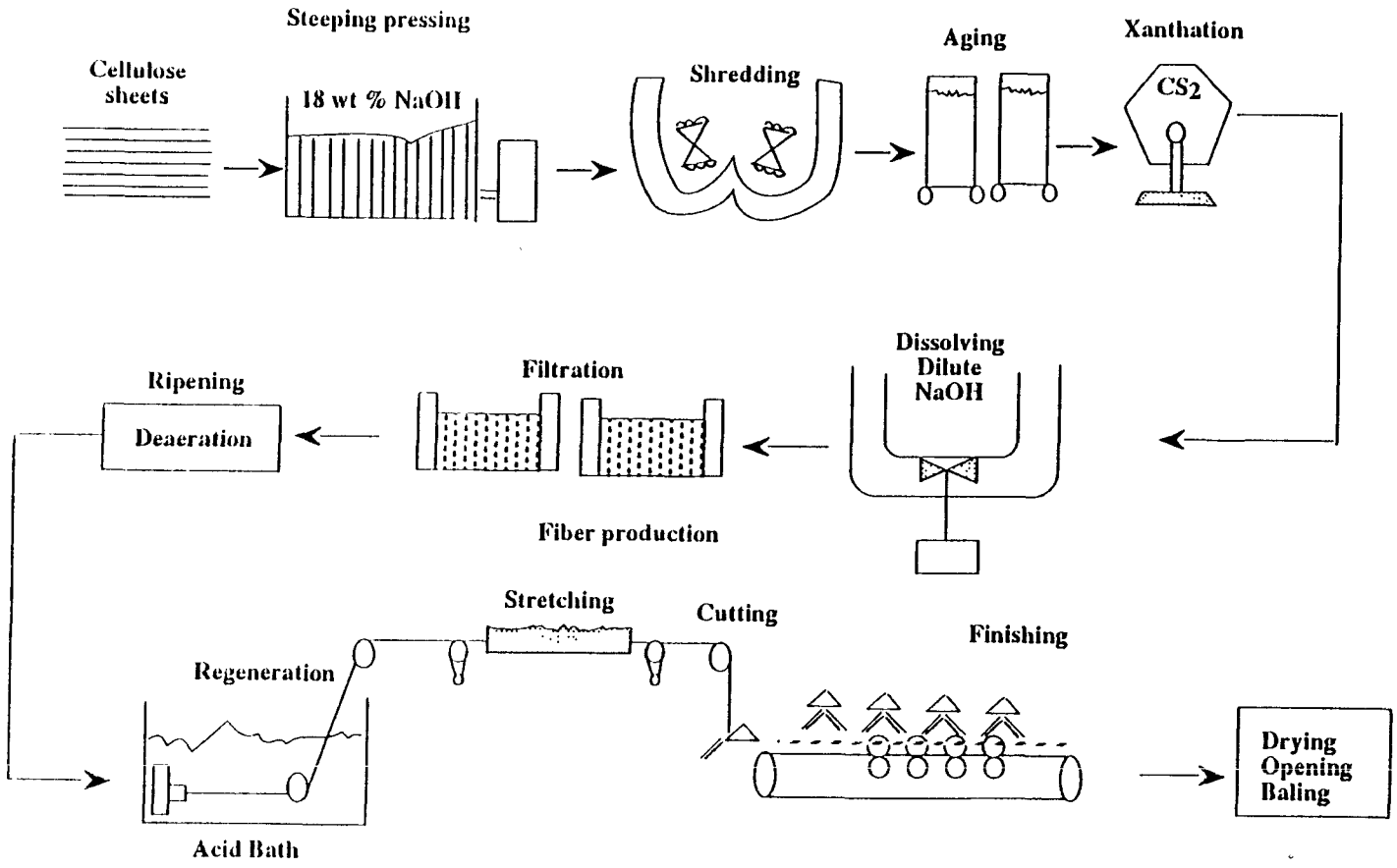
Viscose processes emitted approximately 500 lbs carbon disulfide per ton of rayon fiber produced in 1989 (9). Estimates from 1989 indicate that the viscose process of regenerating cotton and wood cellulose to make rayon and cellophane accounts for about 40 percent of the carbon disulfide consumed in the United States (2). Most of the world's rayon is made by the viscose process (10). In addition to being used to manufacture rayon, cellulose food casings are also manufactured by the viscose process.

Emissions from the viscose process are often high volume/low concentration streams (approximately 400,000 cubic feet per minute (cfm) of water-saturated air containing approximately 100 parts per million (ppm) of carbon disulfide (11)). This can result in difficult and costly emission control problems. Section 4.0 of this report discusses various potential carbon disulfide control options and their limitations.

Process Description

The viscose process used to make rayon is shown in Figure 1 (10). A brief description of the steps involved and those steps wherein emissions can be expected to occur are given below (10, 12).

Figure 1. Viscose Process*



*See Reference 10

Steeping: This step converts cellulose-pulp sheets to alkali cellulose by placing the sheets in a steeping press and filling the press with sodium hydroxide. After steeping, the cellulose is pressed under high pressure.

Shredding: This step is used to convert the alkali-cellulose to a "crumb-like" material. This step also helps distribute the caustic more uniformly in the cellulose while heating the "crumbs" to a temperature favorable for aging.

Aging: Aging is used to decrease and control the degree of polymerization of the cellulose.

Xanthation: This is a critical step in which the alkali-cellulose crumb is reacted with carbon disulfide to form cellulose xanthate. To control the carbon disulfide vapor that escapes, an enclosure system is generally used for xanthation.

Dissolving: This step uses cold dilution in caustic solution to form the cellulose xanthate into a clear, honey-like viscous dope known as viscose.

Ripening: This step allows the clear viscose to coagulate and "ripen" to the proper xanthate level.

Spinning/
Regeneration: In this step, viscose is extruded into a bath containing both salt and acid. The primary goal of the spinning stage is the control of the coagulation versus regeneration rate and the maximal use of the differences in these rates to obtain maximum response to stretching. Carbon disulfide is released during spinning in an open bath.

Stretching: This step allows for better molecular alignment of the viscose fibers through a fixed amount of stretching. Since enclosure is difficult, carbon disulfide is released from the fibers as they are stretched and cut.

Finishing: After the fibers are spun, cut and washed, finishes are applied to give the proper frictional performance in subsequent textile processing.

3.2 Carbon Tetrachloride Production

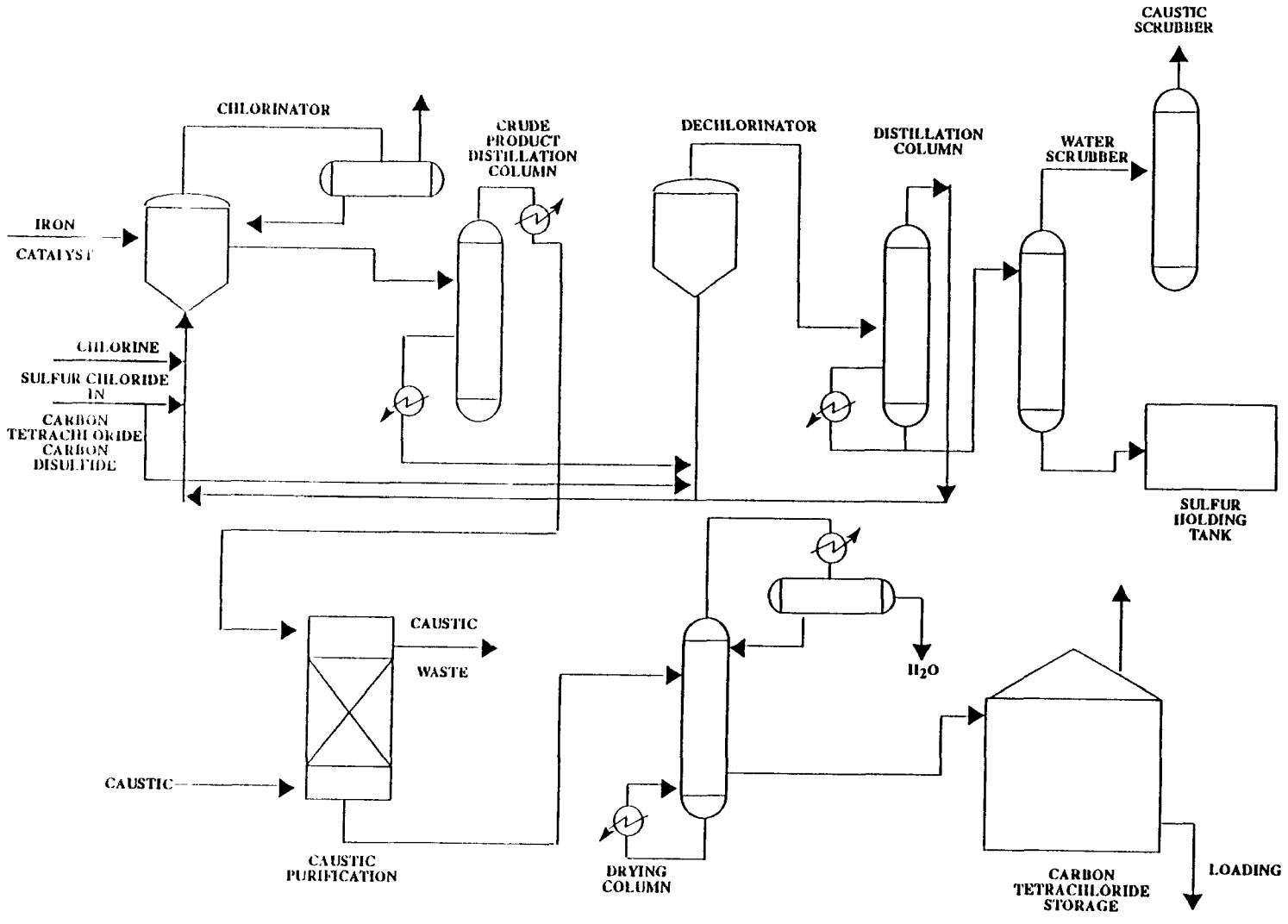
The carbon tetrachloride production process is also an emitter of carbon disulfide. Carbon tetrachloride is produced by the chlorination of carbon disulfide or the chlorination of methane or higher hydrocarbons. Since carbon disulfide is emitted only from the chlorination of carbon disulfide, only that process is discussed. In the carbon disulfide process, a mixture of carbon

disulfide and sulfur monochloride in carbon tetrachloride is chlorinated under controlled conditions. This yields a mixture of carbon tetrachloride and sulfur monochloride, which are then separated by distillation. The sulfur monochloride is recycled. Carbon disulfide is emitted from the chlorinator, the storage facilities, handling (i.e., loading of trucks, tank cars, barges), and as process fugitives. Figure 2 presents a diagram of this process (8).

3.3 Other Processes

Other processes that emit carbon disulfide include the manufacture of thiocyanates and high-purity metal sulfides. Miscellaneous applications include direct uses of carbon disulfide for the cold vulcanization of rubber, as a flame lubricant in cutting glass, and for generating petroleum catalysts. In addition, carbon disulfide is used for dissolving free sulfur, phosphorus, and iodine (2).

Figure 2. Carbon Tetrachloride Production*



*See Reference 8

4.0 CONTROL TECHNIQUES

This section describes control techniques for carbon disulfide. These techniques include existing applications as well as potential applications that are currently being researched. Certain carbon disulfide emission streams are high-volume air streams with low concentrations of carbon disulfide. Some of the control techniques described herein may only be applicable to low-volume air streams with high concentrations of carbon disulfide. Therefore, the nature of the stream needs to be considered when deciding which techniques to apply. These techniques are described for the general case. For a process specific application, consult the particular reference cited. A list of additional references can be found in the October 1988 edition of Pollution Engineering (13).

4.1 Absorption

This subsection outlines the following absorption methods: oil absorption (Methods 1 and 2) and diglycolamine agent absorption (Method 3).

4.1.1 Method 1: Oil Absorption

Vendors of liquid absorption recovery systems claim that carbon disulfide emissions can be controlled by oil absorption systems. The basic process set-up is similar for each vendor in that a combination of carbon disulfide and an absorption liquid are mixed in an absorber tower and then sent to a stripper tower where they are heated and the carbon disulfide flashes out. The nature of the absorption liquid is proprietary information.

The carbon disulfide recovery system operates in two continuous stages. The first stage involves the removal of carbon disulfide from the exhaust stream in an absorber using the absorption liquid. The vapors are absorbed in a counter-flow packed tower absorption vessel. In the second stage the absorption liquid that is now rich with carbon disulfide is passed from the absorber to a series of heat exchangers. This increase in temperature enhances the release

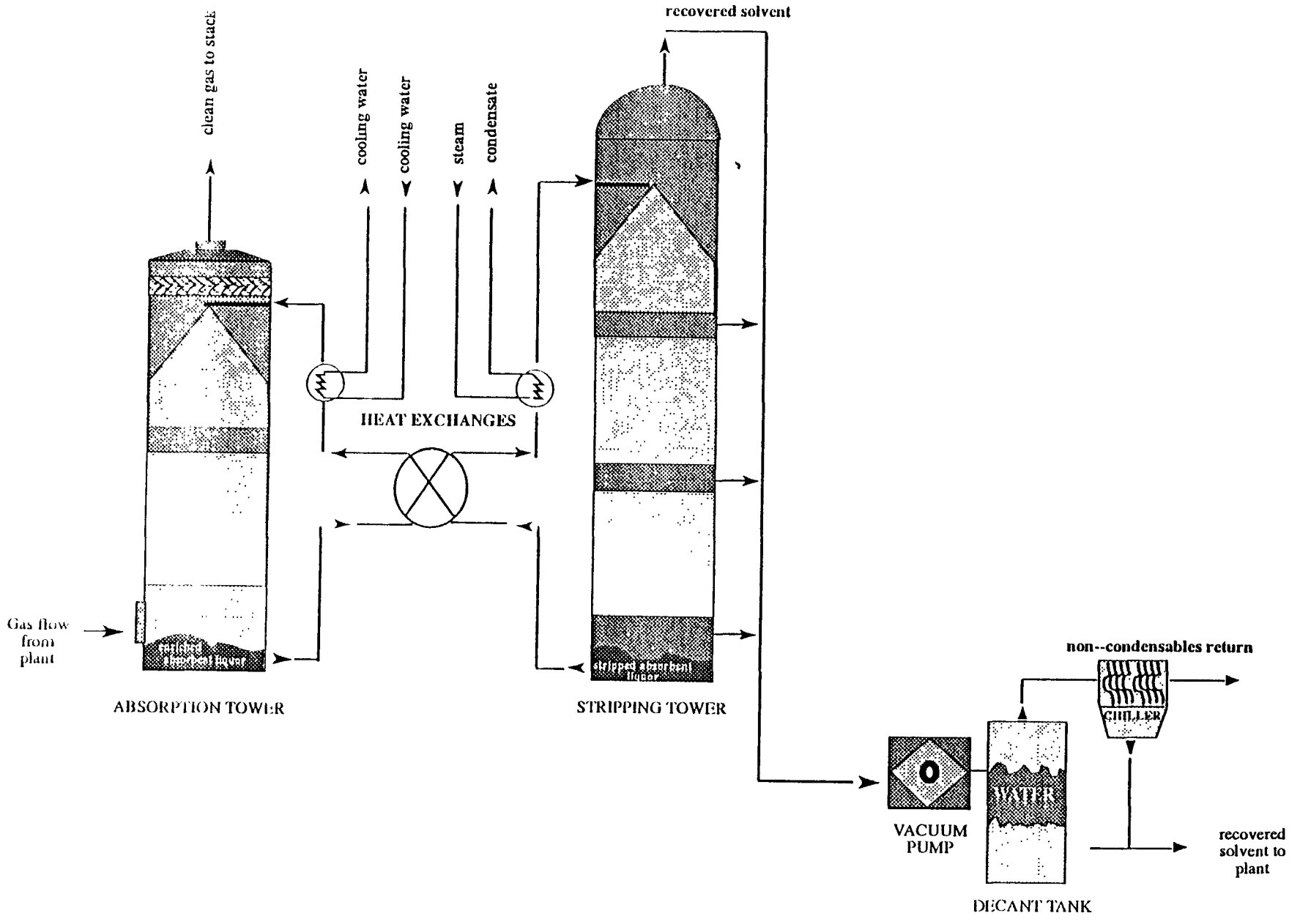
rate of carbon disulfide from the absorption liquid in the stripping column. The stripped absorption liquid is recycled back into the absorber for continuous operation. The separated carbon disulfide vapors are then condensed to the liquid phase. Figure 3 presents a process diagram of this control technique (14). Potential applications for this control technique include textiles, furniture finishing and manufacturing, electronics manufacturing, pulp and paper industries, and petrochemical industries.

There are some limitations to the oil absorption method. Although oil scrubbing may be feasible for emissions with high concentrations of carbon disulfide, a potential concern with this method is that it may not be feasible for emissions with low parts-per-million concentrations. A study of carbon disulfide control options at a cellulose food casing manufacturing facility has shown that gas absorption could be used to remove carbon disulfide from its emissions. However, the recovery of carbon disulfide from the large absorption liquid flow would be economically infeasible. Furthermore, because the absorption liquid could not be regenerated, using gas absorption as the only removal method would be impractical (11). In addition, CMA is concerned that the oil absorption system will require large volumes of oil and that the fugitive emissions of volatile organic compounds from the scrubbing oil will be significant (15).

4.1.2 Method 2: International Patent No. WO 86/02283
Title: Air Scrubbing Process

This technique (16) relates to the purification of air by scrubbing with an effective liquid that can be recycled. This invention presents an air scrubbing process (similar to Method 1) that utilizes a scrubbing liquid which is a complete or partial fatty oil, particularly a drying, semi-drying or saponifiable vegetable, marine, or land animal oil or liquid fat. If the scrubbing liquid is not entirely fatty oil, a substantial portion of the remainder of the scrubbing liquid is water with which the oil or fat forms an emulsion. The scrubbing liquid can also include a small proportion of a detergent or wetting agent.

Figure 3. Oil Absorption System*



*See Reference 14

According to the patent, carbon disulfide is one of the many pollutants that may be purified from an air stream using this process. After treatment of the air, the scrubbing liquid can be precipitated readily from the air stream so that no appreciable amount of the scrubbing liquid will be lost in the air discharged from the treatment process. The patent also states that the scrubbing liquid is readily available and inexpensive (16). The reader should be aware that this air scrubbing process is an experimental process and has not yet been demonstrated to be effective in an industrial setting.

Selection of Scrubbing Liquid

The following fatty oils are suggested for use for the scrubbing liquid because of their ready availability and effectiveness.

soybean oil
cottonseed oil
groundnut or peanut oil
corn oil
safflower oil
sunflower seed oil
rapeseed oil

Reference 16 contains an extensive listing of other oils that may be used.

In order to facilitate spraying and dispersion of the scrubbing liquid containing fatty oil, it is preferred that the oil be emulsified with water to provide a scrubbing liquid with a viscosity lower than the viscosity of the oil. In order to condition the oil for reuse, the oil and water are separated and pollutants removed from the air are stripped from the oil or fat. Such stripping can be effected by the use of steam in accordance with known procedures. Steam stripping of fatty oils is disclosed in Wecker U.S. Patent 1,622,126 (1927) (17). Stripping of fatty oils can also be accomplished continuously by use of the deodorization procedure disclosed in Dean U.S. Patent 2,280,896 (1942)(18). The reader is encouraged to locate these patents if this control technique is considered.

4.1.3 Method 3: Diglycolamine Agent

Diglycolamine (DGA) agent is manufactured by Texaco Chemical Company and is the brand name for 2-(2-aminoethoxy)-ethanol. It was first introduced in the United States in 1965 and is primarily used for removal of hydrogen sulfide and/or carbon dioxide from gas streams. DGA agent is a clear, viscous liquid at room temperature and a white crystalline solid below its freezing point of 9.5° F (19). Although DGA agent is used primarily for hydrogen sulfide and carbon dioxide absorption, it may be possible to use DGA agent to control carbon disulfide emissions through a gas absorption system as discussed in Method 1 (19). Although there are no existing applications of this technology for the purpose of carbon disulfide control, it may still be considered as a potential control. DGA agent reacts with carbon disulfide to produce the major degradation product N,N'-bis(hydroxyethoxyethyl) urea. The chemical equation for this reversible reaction is

$$2R{-}NH_2 + CS_2 \overset{\Delta}{\rightleftharpoons} R{-}\overset{\overset{H}{|}}{N}{-}\overset{\overset{S}{\|}}{C}{-}\overset{\overset{H}{|}}{N}{-}R + H_2S$$

where

$$R = HO{-}CH_2{-}CH_2{-}O{-}CH_2{-}CH_2$$

This degradation product has a much lower toxicity than the parent compound, carbon disulfide (19).

4.1.4 Conclusions

Absorption may be a feasible control technique for high-concentration, low-volume carbon disulfide air emissions. However, it may not be feasible for low-concentration streams. It is recommended that the reader further research the absorption methods presented herein prior to considering using these methods. The selection of the absorption medium is critical to the

success of the control technique. The preceding sections have identified absorption liquids that are thought to be effective in removing and/or eliminating carbon disulfide. According to some industrial contacts, there are certain problems with using oil absorption in that it is not as effective as vendors claim. Also, gas absorption is equipment intensive and for that reason may be more costly than other alternatives. For example, a study conducted by Argonne National Laboratory showed that application of conventional gas absorption processes for carbon disulfide removal is relatively expensive mainly due to low equilibrium carbon disulfide loadings in all possible absorbents. There are requirements for high liquid flow and relatively low superficial gas velocities to avoid flooding in absorption towers. From this study it was determined that 13 conventional absorption towers (12 ft in diameter) would be required for carbon disulfide removal for a stream with a 100 ppm carbon disulfide concentration (11). Pilot plants studies or trial plant runs may be required to determine effectiveness and suitability of this control technique to a particular carbon disulfide stream.

4.2 Adsorption

This subsection presents adsorption methods using activated carbon and a synthesized sorbent technique.

4.2.1 Method 1: Activated Carbon

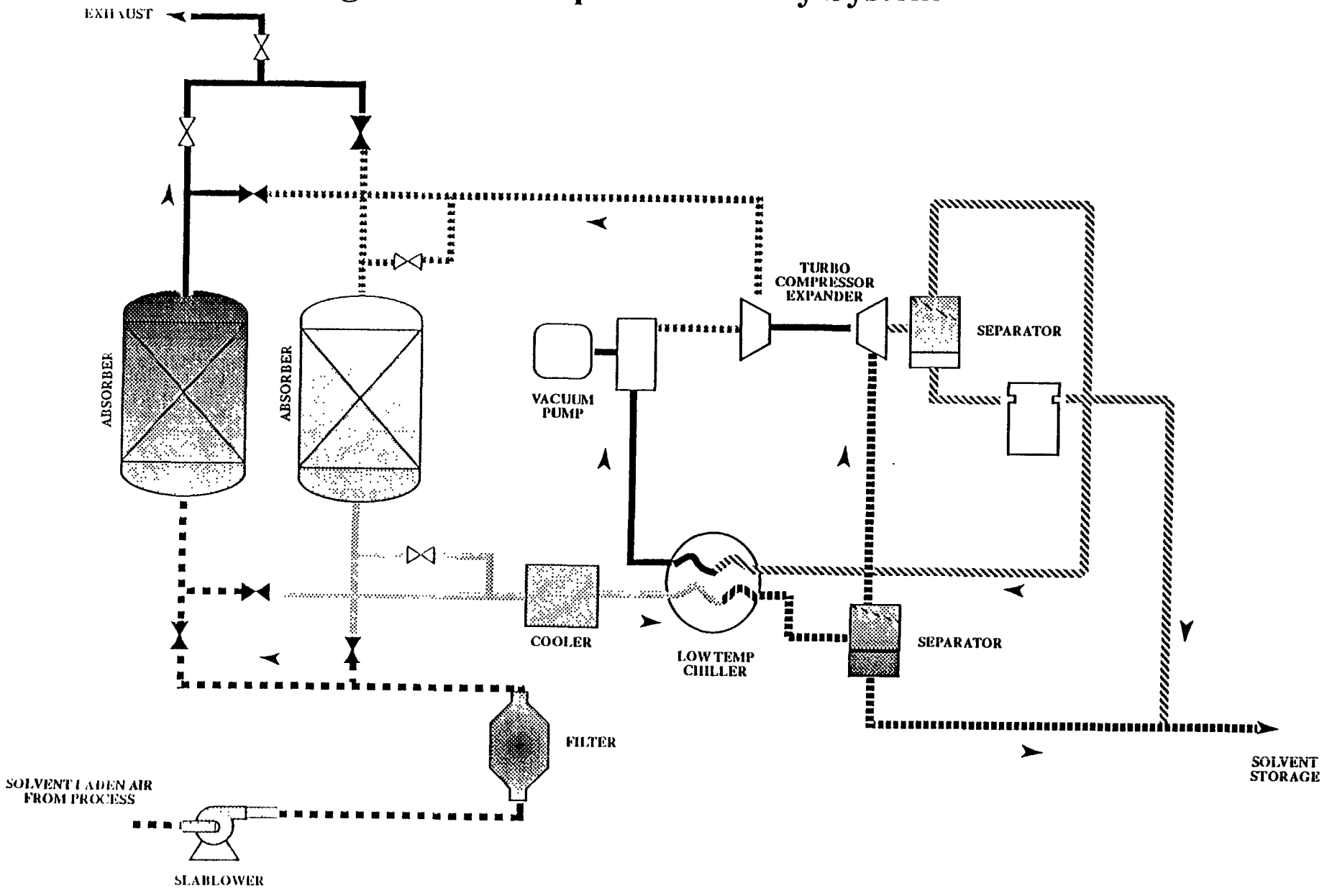
Carbon adsorption is a traditional control technique used for solvent recovery, energy recovery, odor removal, and regulatory compliance. This control technique employs the use of activated carbon as an adsorption medium. Air contaminated with carbon disulfide is passed through a bed of the activated carbon. Carbon disulfide is adsorbed onto the internal surface of the adsorbent and the purified air is exhausted to the atmosphere. For most applications, the adsorbent bed is then regenerated, generally with steam. In terms of carbon disulfide adsorption, the primary problem associated with steam regeneration is the possibility of fires since the autoignition temperature of carbon disulfide is below the temperature of steam. The carbon disulfide is then extracted from the steam through condensation.

One method of eliminating the fire problem associated with steam regeneration is to use dry nitrogen for regeneration. In this application, hot, inert gas (nitrogen) is passed through the adsorbent bed and desorbs the adsorbed carbon disulfide. The carbon disulfide-rich inert gas is cooled and passed through the compressor side of a turbo unit. It is further cooled in an interchanger and enters the expansion side where cooling as low as -80 °F is accomplished. Condensed liquid is separated and the inert gas is passed through a mechanical pump and is returned to the adsorber. Figure 4 presents a complete process diagram of this recovery system (20).

There are safety concerns associated with the use of activated carbon to control carbon disulfide emissions. Carbon disulfide differs from organic compounds typically treated by activated carbon systems. Organic compounds typically controlled by carbon adsorption have an explosion hazard classification of "slight" or "moderate", whereas carbon disulfide has a "severe" explosion hazard classification. In addition, carbon disulfide exhibits a much lower flash point than typical organic solvents treated by activated carbon systems. Air and/or rust are listed as "incompatible materials" for carbon disulfide but are not considered incompatible for conventional solvents handled by activated carbon systems (21). Carbon disulfide reaction products of activated carbon systems are much more corrosive than for typical volatile organic compounds. Sulfuric acid is produced by the contact of reduced sulfur compounds on activated carbon systems. Furthermore, the presence of sulfuric acid requires extensive custom design and exotic metallurgy for pipes, condensers, heat exchangers, valves, etc. to control corrosion (21).

In addition to the above concerns, carbon disulfide exhibits a much higher vapor pressure than typical organic solvents. At stack conditions, carbon disulfide vapor pressure is approximately 450 mm Hg; at 25°C it is 350 mm Hg. Vapor pressure of typical organic solvents range from 2 mm Hg to 76 mm Hg. The adverse effect of a high vapor pressure is that condensation of carbon disulfide would require extensive refrigeration rather than conventional water cooling to condense the vapor after column stripping (21).

Figure 4. Adsorption Recovery System*



*See Reference 20

Studies performed by Argonne National Laboratory using various activated carbons for carbon disulfide removal indicated that this method is definitely superior to other methods for removal and recovery of carbon disulfide from air. Carbon has a great affinity for carbon disulfide. It was concluded that activated carbon may make efficient adsorption and recovery possible if the other known problems (H_2S and H_2O poisoning, water loading, and fire hazard) can be overcome (11). Further study of the carbon adsorption method was recommended by Argonne National Laboratory.

4.2.2 Method 2: Synthesized Sorbent: European Patent EP 189606

This patent (22) discusses an invention that involves impregnating a solid material suitable for selective and quantitative removal of carbon disulfide. Some of the solids that can be used for the impregnation treatment are silica gel, alumina, clay mineral and zeolites. The active agents for the impregnation procedure which have been found to be effective are amines and amine complexes of their derivatives. For the impregnation procedure, the solid is mixed homogeneously with the active agents at temperatures below 200 °C. The loading of the solid material can be controlled by the initial amount of the active agent in contact with the solid. Afterward, the mixture can be thermally treated (below 300 °C) and dried in air or in an inert atmosphere. This thermal treatment determines the ultimate loading of the active agent on the solid. After a reconditioning of the impregnated material (flow of dry inert gas), the material is activated. This creates high selectivity for the impregnated solid for adsorption of carbon disulfide. The adsorbed carbon disulfide can then be quantitated down to the ppb range with a universal S-monitor (22).

The patent described above is only a potential option that should be further researched before being used. At this point, the synthesized sorbent method is <u>experimental technology</u>.

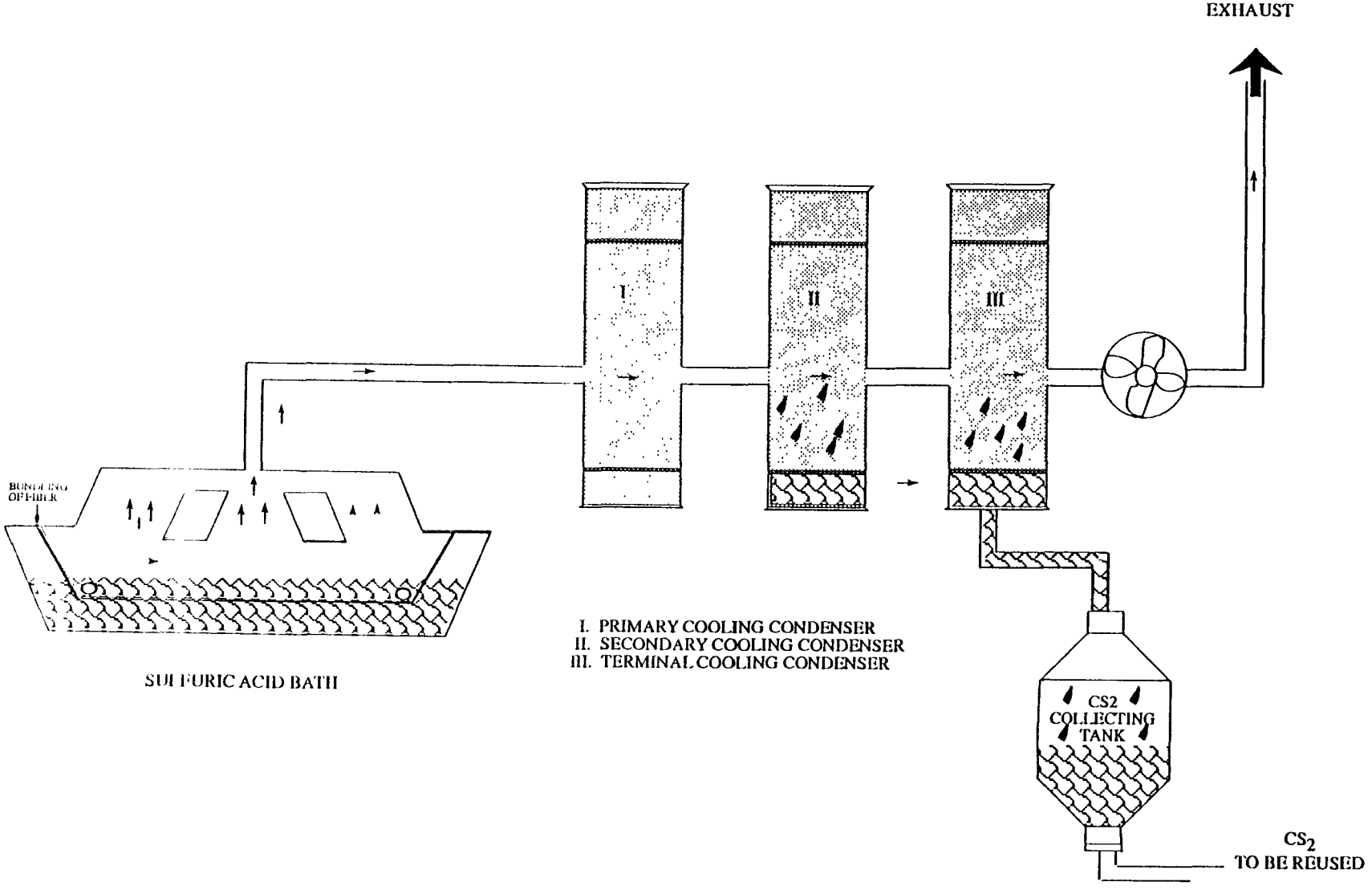
4.2.3 Conclusions

Adsorption is a traditional and feasible control technique and is relatively easy to operate. However, for carbon disulfide, it is imperative that steam is not used in regeneration due to the relatively low autoignition temperature of carbon disulfide. Some other regeneration alternatives have been presented (i.e., nitrogen desorption). Although nitrogen desorption solves the problems of using steam, it introduces additional problems (i.e., storage, handling). Carbon adsorption, as well as the newer synthesized sorbent process, present beneficial, and potentially useful, control technologies for carbon disulfide emissions. As previously mentioned, Argonne National Laboratory determined that activated carbon may make efficient adsorption and recovery possible.

4.3 Ventilation and Condensation Recovery System

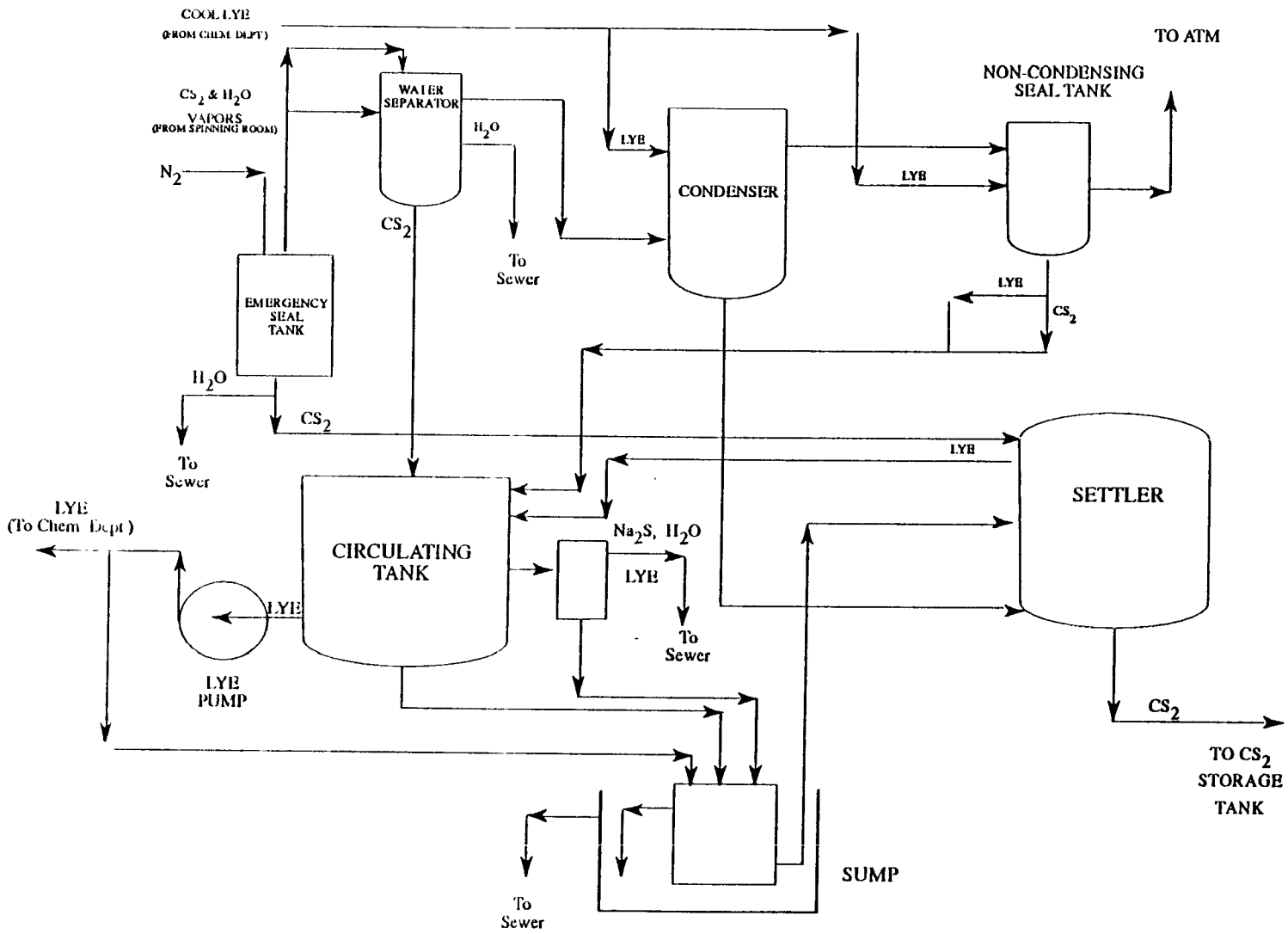
A ventilation and recovery system is used by the Shanghai Number 12 Viscose Rayon Factory (12). Since 1972, carbon disulfide vapors from the spinning process (see Section 3.1) have been controlled by a system of exhaust ventilation and recovery. The recovery of carbon disulfide is achieved after the capture of the vapor in the ventilation pipe by three successive cooling condensers and a collection system. Figure 5 presents a schematic of this control technique (12). During the early 1980s, the average rate of recovery (efficiency) reached 48 percent of all carbon disulfide added in production. A similar carbon disulfide control/recovery system has been in operation at the BASF Corporation, Fibers Division, Lowland, TN, since 1956 (23). This system employs carbon disulfide condensation through a cold caustic solution and carbon disulfide recovery in a receiving tank. It works most efficiently when carbon disulfide concentrations are at very high levels in a low-volume stream. The collected carbon disulfide is then transferred to a storage chamber as pure carbon disulfide. Eleven to twelve percent total carbon disulfide is recovered for reuse. An additional 20-30 percent of the carbon disulfide is converted to hydrogen sulfide. Figure 6 shows a diagram of the carbon disulfide recovery system used by BASF (24). Recovery techniques are applied by BASF to high concentration areas of the spinning-process instead of to the low concentration high volume exhaust stream at the final steps of the process (24).

Figure 5. Ventilation & Recovery System*



*See Reference 12

Figure 6. BASF Carbon Disulfide Recovery System*



*See Reference 24

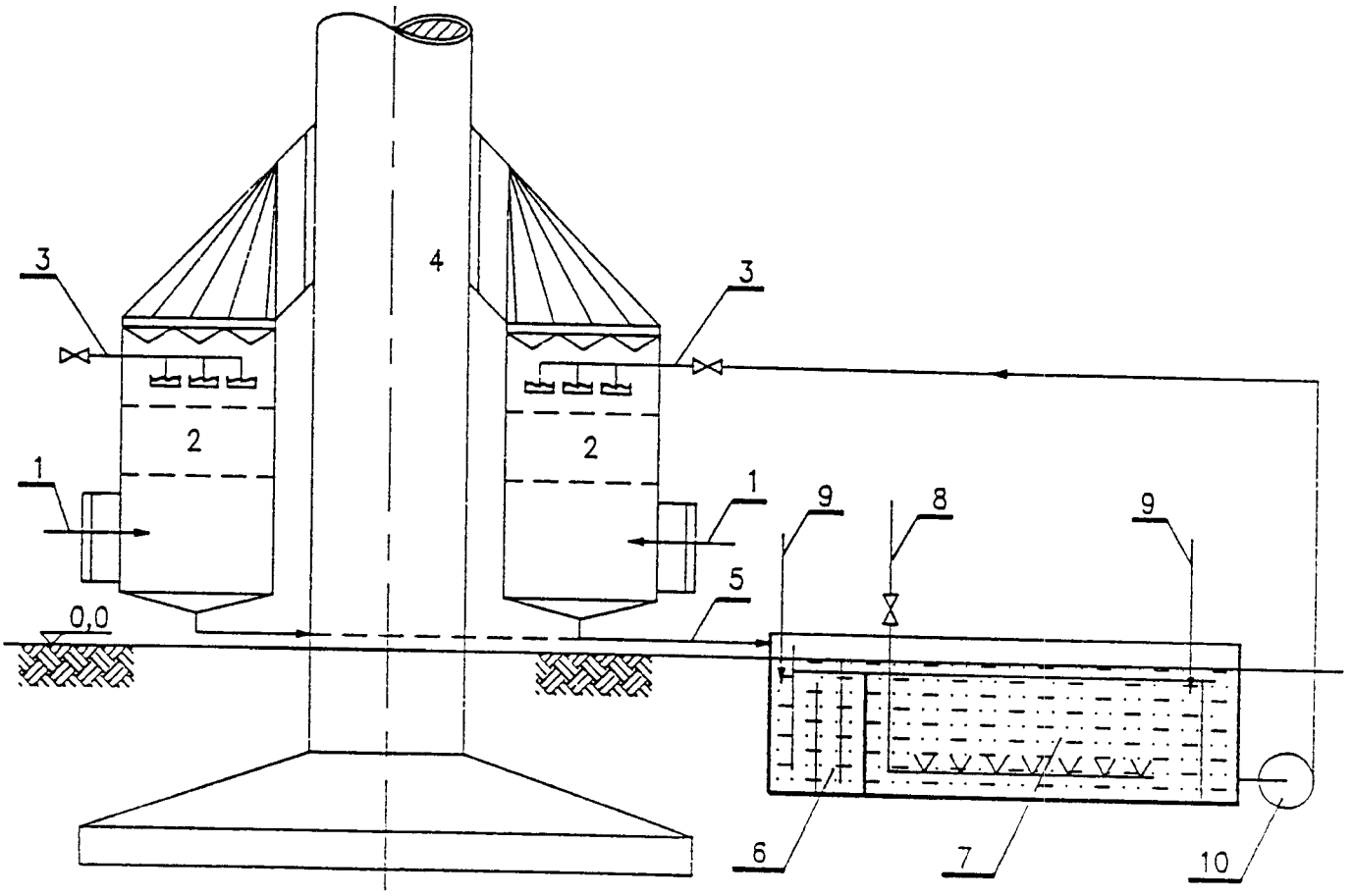
This particular control technique has already been applied at several existing sites. It can be concluded that it is a feasible option for high concentration streams (15). It should be noted, however, that this arrangement was designed specifically for the rayon production process and may not be suitable for other sources. It should be noted that refrigerated condensers are currently being used in a carbon disulfide production process (25); however, this technique poses a disposal problem if the carbon disulfide is not to be recycled back into the process.

Ventilation and recovery systems could involve prohibitively large energy requirements for cooling. For example, cooling the enormous volumes of water-saturated air typical of viscose plant exhaust would be economically and technically infeasible for low concentration carbon disulfide emission streams (15). However, condensation technology is an option that may be considered for areas where emission streams with very high carbon disulfide concentrations can be captured.

4.4 Absorption/Oxidation: U.S. Patent 4,049,775

The method discussed in the patent, developed at the Institute of Synthetic Fibers, Lodz, Poland, is experimental and is being tested on an industrial scale at CHEMITEX-WISTOM Synthetic Fibre Industry Plant, Tomaszow Mazowiecki, Poland (26). This patent discusses a process that removes hydrogen sulfide, carbon disulfide, and sulfur dioxide from the polluted air in one operation by absorbing and oxidizing the gases in a bath. The specific installation discussed in the patent is used to remove hydrogen sulfide and carbon disulfide from the ventilation system air in viscose fiber plants. The installation has a capacity of one million cubic meters per hour and an alkaline bath (pH= 9.6 - 9.8) with a redox system being used for absorption purposes. The carbon disulfide is absorbed by the bath in the presence of hydrogen sulfide and is subjected to hydrolysis. The carbon disulfide reaction and hydrogen sulfide absorption products are oxidized with atmospheric air to sodium thiosulfate, elemental sulfur and a small amount of sulfates. This technology can reduce carbon disulfide concentrations of 200-600 mg carbon disulfide per cubic meter by about 70 percent. Figure 7 presents the process description for this technique (26).

Figure 7. Wet Method of Carbon Disulfide Control*



A schematic drawing of the installation for removing CS_2 and H_2S from the ventilation system air in viscose fibre plants: 1–inlet of gases into the towers, 2–plate columns, 3–reflux inlet, 4–scattering chimney 150 m high, 5–discharge of bath solution from the towers, 6–bath solution seasoning chamber, 7–aeration reservoir, 8–sparger air inlet, 9–bath solution components' inlet, 10–reflux pumps

*See Reference 26

The plant equipment is comprised of 5 towers with 7.8 meter diameter plates. The cylindrical section of a 5 meter high tower ends in a 5 meter high tapered section feeding the gases into the chimney. A liquid condenser is positioned between the cylindrical and tapered sections. The gases are fed into the tower by a fan that delivers from 250,000 to 300,000 cubic meters per hour, which is equivalent to an average flow rate of gases through the tower of between 1.5 to 1.8 meters per second. The absorption bath is pumped from the reservoirs situated below ground level. The bath solution flows down the towers into the reservoirs where it is regenerated. Bath regeneration is achieved by subjecting it to oxidation with air. The air is fed by compressors with an output of 1650 cubic meters per hour. The purified air is emitted to the atmosphere by a scattering chimney 150 meters high (26).

As this is a foreign installation, it is difficult to judge its feasibility for a domestic application. As previously stated, this technology is currently experimental. The patent does claim a 70 percent reduction of carbon disulfide emissions and an emission rate of 10^6 m^3 carbon disulfide/hour. Further investigation into the nature of the bath and its effectiveness for carbon disulfide is recommended.

4.5 Regenerative Thermal Oxidation

This control technique involves incineration of the gas stream containing carbon disulfide. The basic construction for this control option admits process gas into the thermal oxidation unit through an inlet manifold. A flow control valve directs this gas into an energy recovery chamber in inlet (preheat) mode. The process gas and contaminants are progressively heated in the stoneware bed as they move toward the purification chamber. High temperature thermal oxidation is accomplished in this central chamber. Gas or oil burners maintain a preset destruction temperature of up to 1100°C (2012°F) with a residence time of up to one second. Carbon disulfide present in the gas stream will autoignite, releasing energy in the stoneware bed.

From the central chamber, the purified process gas stream passes through an energy recovery in outlet (heat recovery) mode. The cleaned gas can be handled by a standard fan

located in the clean gas side. Figure 8 presents a schematic of this technique (27). Several applications for this process include chemical processing; metal coating; petrochemical industries; pulp and paper industries; and paint, solvent, and ink manufacturing.

According to equipment vendors, this technique is feasible and has many applications for industry pollutant control. However, it may be cost prohibitive as the process equipment would need to be made of Hastalloy or Stainless Steel due to the corrosive nature of carbon disulfide. In addition, substantial energy costs are associated with using this option for viscose processes (15). Another potential drawback of using thermal oxidation is that sulfur dioxide is an oxidation by-product of carbon disulfide. Control of the resulting sulfur dioxide emissions is a concern.

4.6 Catalytic Conversion: U.S. Patent 4,374,819

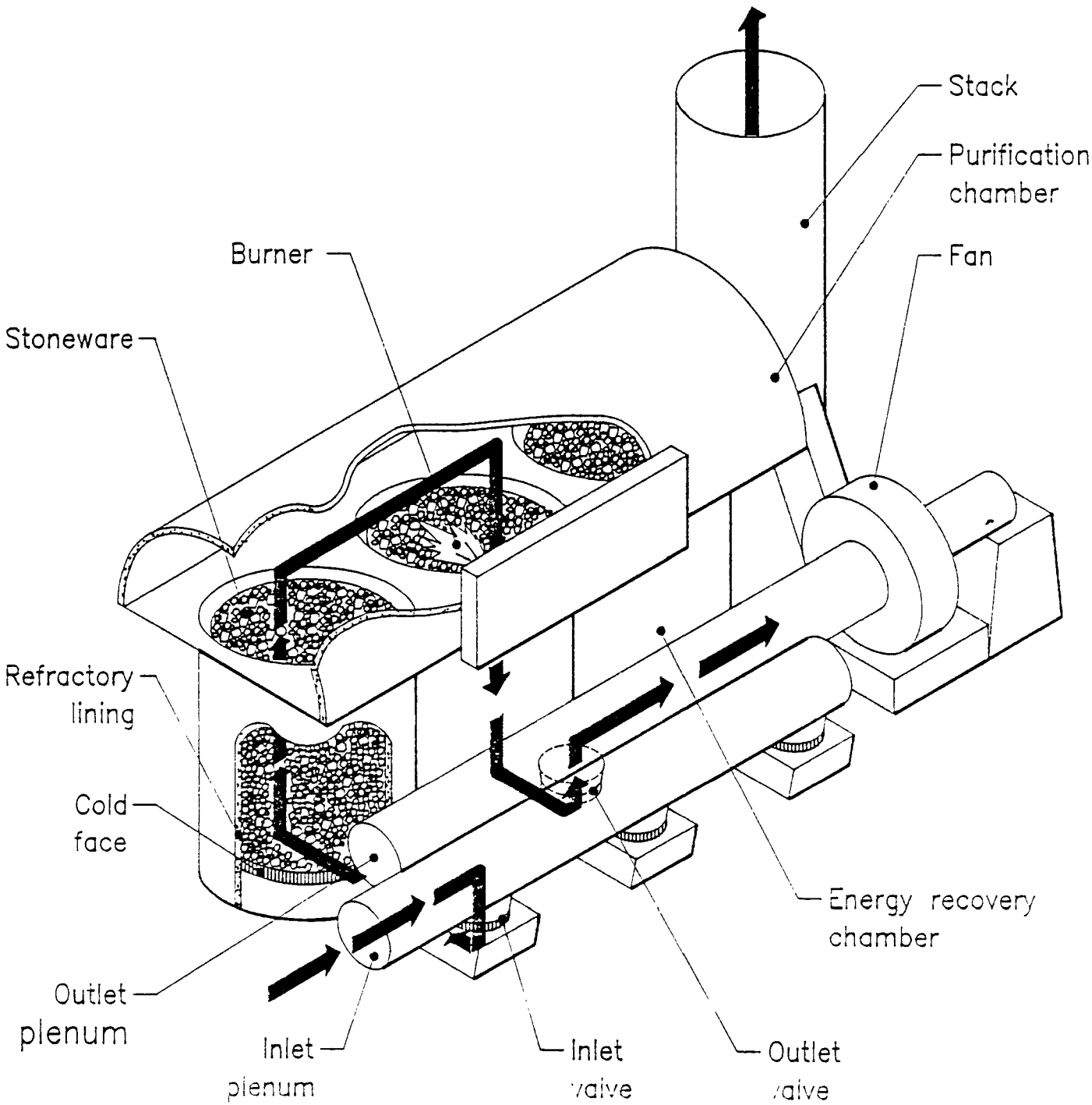
This patent discussed a system that relates to a multi-stage process for removing noxious sulfur containing gases, notably hydrogen sulfide, sulfur dioxide, carbonyl sulfide and carbon disulfide, from waste gas streams (28). This control technique employs the Claus reaction for sulfur recovery:

$$2H_2S + SO_2 \rightarrow 2H_2O + 3/2\ S_2$$

The Claus process has been used to remove hydrogen sulfide from gas streams by reacting hydrogen sulfide with sulfur dioxide in the presence of a catalyst (i.e., bauxite, alumina, cobalt molybdates) (28). The first stage involves a Claus reaction, but uses a novel catalyst which also permits the reduction and/or elimination of the carbon disulfide from the gas stream through the following reaction:

$$CS_2 + SO_2 \rightarrow S_x + CO_2$$

Figure 8. Thermal Oxidation System*



*See Reference 27

In this first stage, the gas stream is passed through a reactive zone having a temperature between 150 °C to 350 °C in the presence of a pre-treated novel catalyst having the following formula:

$$xLn_2O_3yT_2O_3$$

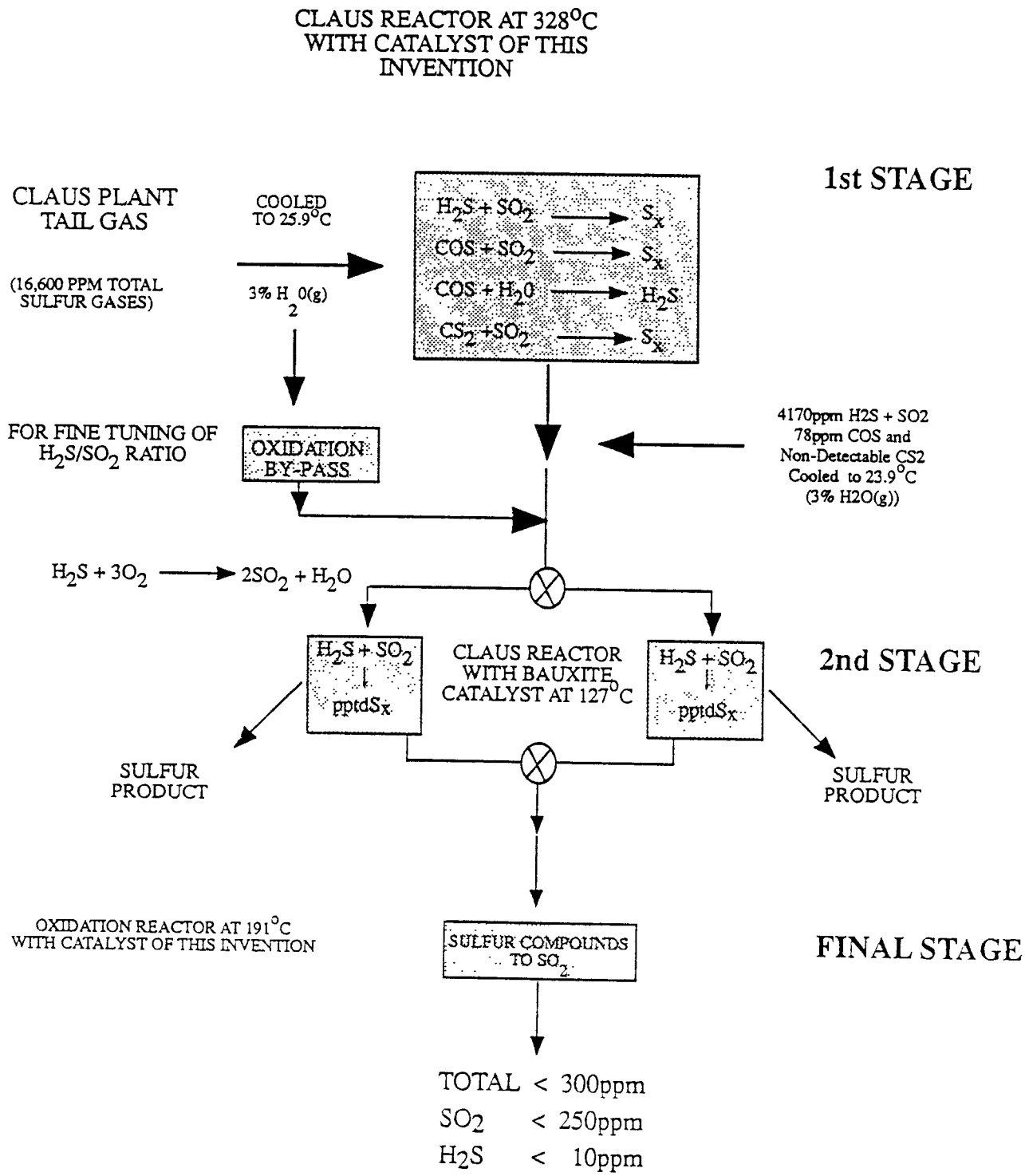
In this formula, Ln is yttrium or a rare earth element, T is cobalt, iron or nickel, and x and y are independently numbered from 0 to 3. The catalyst is non-crystalline and has a surface area of about 10 m^2/g to about 40 m^2/g. The preferred catalyst is one in which Ln is lanthanum; T is cobalt; and x and y range from 1 to 3, including non-integers. The first stage yields a product stream having a reduced content of sulfur-containing gases, specifically including substantial reduction and/or virtual elimination of carbon disulfide.

An intermediate stage is a Claus reaction which may take place in one or more reaction zones in the presence of known catalysts such as bauxite, alumina or cobalt molybdates. The final stage is required to reduce the hydrogen sulfide concentration to about 10 ppm. This stage involves the air oxidation of hydrogen sulfide at a temperature between 150 °C to 300°C in the presence of a catalyst usable in the first stage. Figure 9 presents a diagram of the three stages (28). For carbon disulfide removal, only the first stage needs to be executed.

In general, this technology applies to commercial processes involving very high concentrations of sulfur gases. It is used primarily to control hydrogen sulfide emissions from sulfur recovery plants associated with oil refining operations. The process gases are characterized as containing 15 percent to 20 percent carbon dioxide and essentially no oxygen (15).

This option may also be very costly because the equipment must be constructed of Hastalloy or Stainless Steel. The other primary problem with this option is that the carbon disulfide is not recoverable. If the particular process requires a recycle stream of carbon disulfide, this control option would not be feasible. Although theoretically it appears that this

Figure 9. Catalytic Conversion*



*See Reference 23

method may be used to control carbon disulfide emissions, there have not been any proven practical applications yet.

4.7 Control with a Bioreactor

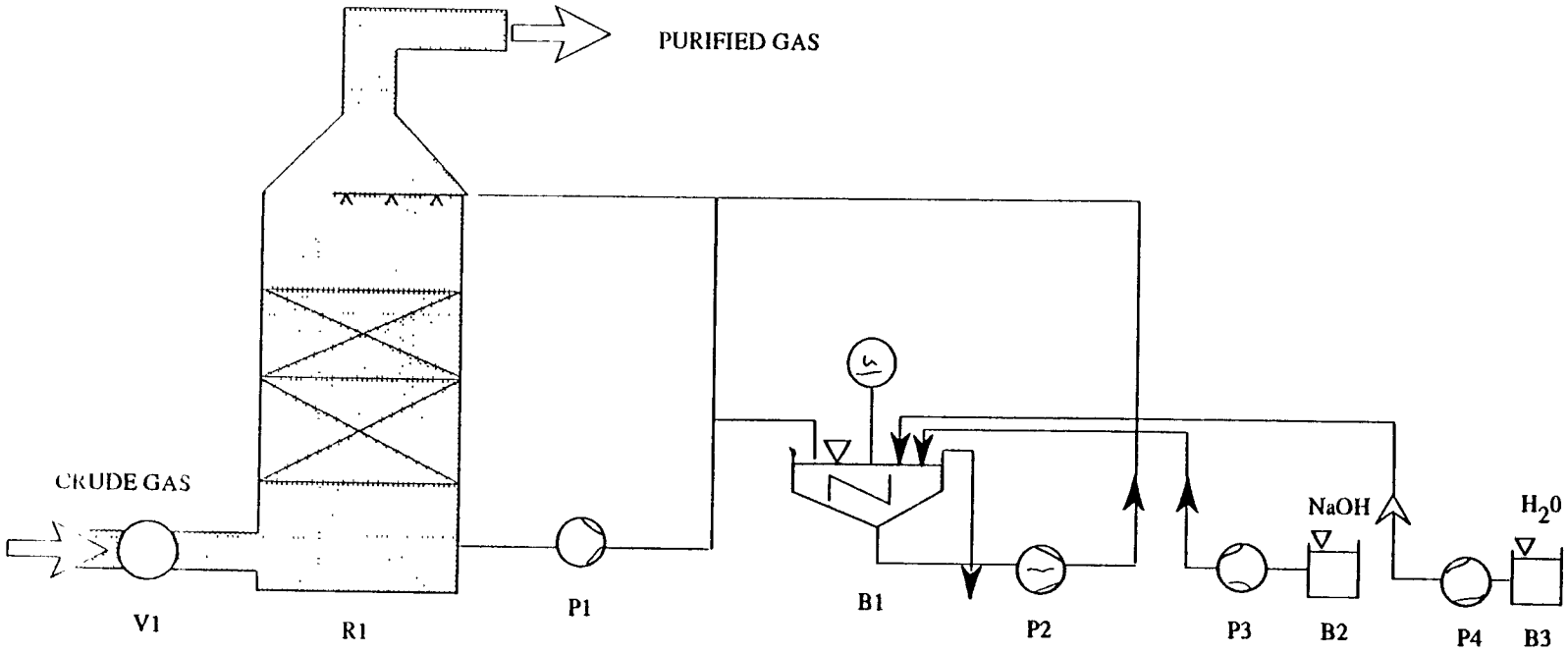
This method, entitled the Waagner-Bio/Glanzstoff Austria Process, employs a fixed bed bioreactor that is designed as a counter current gas-liquid reactor. The reactor is equipped with commercially available packing materials such as polyethylene and polypropylene of different specific areas. The reactor is also equipped with an immobilized mixed culture of microorganisms, generally Thiobacillaceae. The packing material carries the immobilized microorganisms. The gas containing the contaminants (i.e., carbon disulfide) flows counter-current to the water/activated sludge suspension through the reactor. As the crude gas passes through the reactor, the hydrogen sulfide and carbon disulfide transfer into the liquid surrounding the cells and then further through the microorganism cell walls due to the favored rapid oxidation of the hydrogen sulfide and carbon disulfide by the metabolism of the sulfur-oxidizing microorganisms. The oxidation products are released through the cell walls into the liquid and are removed continuously by the circulating flow of the water/activated sludge suspension and are neutralized by the addition of sodium hydroxide. The formed sulfate is removed from the settler by the continuous addition of water. Figure 10 presents a process diagram of this method (29).

This control option has not been proven on an industrial scale, and all data are from pilot plant studies. Further investigation into the capability of this option on actual industrial sites is needed. Theoretically, this option appears to be feasible. However, there is concern that this technology may not be feasible for low concentration, high volume streams (15).

4.8 Wet Scrubbing

Based on more than a decade of wet scrubbing experience, an odor control system has been developed that treats air flows from 500 to 70,000 cfm (30). This method uses a packed

Figure 10. Control With a Bioreactor*



V1 blower
R1 bioreactor
P1 circulation pump
B1 sedimenter
P2 reflux pump
P3 pump for alkali
B2 alkali tank
P4 tap water pump
B3 tap water tank

*See Reference 29

tower concept. In operation, scrubbing spray is created by controlled mixing of sodium hypochlorite (bleach) and/or other chemicals with tap water that has been pretreated to remove hardness. The mixture is then fed to a nozzle located at the inlet of each reaction tower. While odorous air is vented into a reaction tower, the nozzle sprays a fog of very fine droplets which travel concurrent with the air. The nozzle produces droplets with mean diameters of 10-12 microns, which are ideal for effective odor removal. Enhanced by long gas-liquid contact times (typically 10 to 30 seconds), the small fog droplets achieve optimal contact. As a result, odor molecules are completely reacted and clean air is exhausted to the atmosphere. The control mechanism for this process involves non-liquid absorption and subsequent condensation in a packed tower.

A cellulose food casing facility tested this wet scrubbing method at its facility for carbon disulfide removal and determined that it was not feasible on its air stream (31). However, this wet scrubbing method may have the potential to control carbon disulfide emissions upon further study. This method is currently employed at more than 65 wastewater treatment plants in the United States (30). Figure 11 presents a diagram of the control system (30).

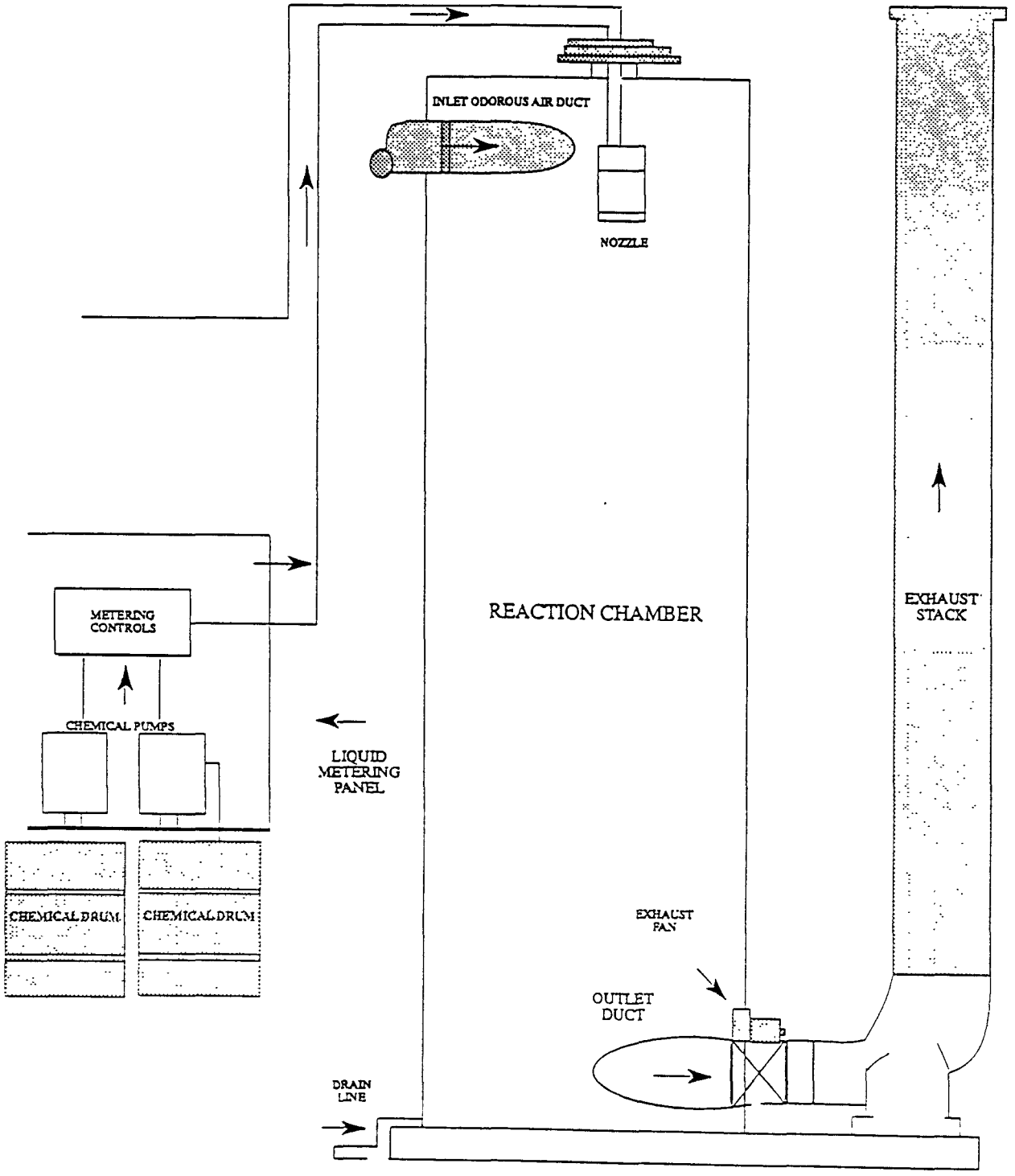
4.9 Current Research

Researchers at Argonne National Laboratory, in Chicago, Illinois, in conjunction with the Illinois Department of Energy and the Department of Commerce and Community, have examined the following four possible options for controlling carbon disulfide emissions.

1. gas absorption
2. gas adsorption
3. membrane separation
4. catalytic conversion

Options 1, 2 and 4 have been presented in Sections 4.1, 4.2, and 4.6, respectively. Option 3, membrane separation, involves the separation of a gas or liquid through the use of a permeable membrane. The driving force for transport is either pressure or concentration (32).

Figure 11. Wet Scrubbing*



*See Reference 30

The following conclusions have been reached concerning the above carbon disulfide control options based on studies conducted at a cellulose food casing facility. Gas absorption was determined to be relatively expensive and was concluded to be infeasible. Results of studies showed that although ways to alleviate the fire hazard danger must be developed, carbon adsorption presents the best current possibility for carbon disulfide recovery. Pursuing the membrane separation option is not recommended at this time because it was determined that existing ceramic membranes cannot remove carbon disulfide from air effectively, even at high carbon disulfide emissions. Finally, it was determined that because the catalytic incineration process would require a catalytic reactor to convert sulfur dioxide to sulfur trioxide and a sulfuric acid plant, there is little economic incentive to choose this option for carbon disulfide emission control (11).

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16. ABSTRACT

The EPA Control Technology Center, OAQPS, has received numerous inquires regarding control options for carbon disulfide emissions. This report presents a description of methods or techniques used to control carbon disulfide emissions from a variety of source categories, and discusses the feasibility of application of each technique presented. This report briefly discusses various options available to public and private industry for controlling carbon disulfide emissions, and serves as a reference source for those seeking further information.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
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Emissions Control Technologies		
Emission Source		
Pollutant Control		

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