Cyanides have been shown to be extremely mobile in the soil environment and have been shown to move from soils to groundwater. Thus, cyanide is also available for potential release and transport to environmental receptors. Cyanide has been found in several different waste streams from coal technologies.

Significant levels of chromium have been detected in solid residues from various coal processes. Table 7.3 shows the chromium concentrations in leachate from certain wastes from primary plating processes. These concentrations may be comparable to those found in leachate from coal ash.

## 7.4 CREOSOTE PRODUCTION INDUSTRY

The EPA has determined that solid wastes from creosote production may pose a substantial present or potential hazard to human health or the environment because of the following considerations:[109]

1. The hazardous substances likely to be present in the wastes include creosote and its constituents, benz (a)anthracene, benzo(b)fluoranthene, and benzo(a) pyrene, all of which are carcinogens. Several reported cases of cancer in humans have been attributed to creosote exposure.
2. If the lagooning/landfilling of these wastes is improperly conducted, the contamination of soil, land, and ground

Table 7.3 Levels of Chromium (mg/L) in Leachate from Electroplating Sludges and Coal Ash

| | |
|---|---|
| Primary Plating Process[a] | |
| Segregated Zinc | 1.22 |
| Multiprocess Job Shop | 0.2 |
| Multiprocess with Barrel or Vibratory Finish | 0.32 |
| Printed Circuits | 0.12 |
| Coal Ash Leachate[b] | 0.014 - 1.0 |
| Water Quality Criteria | |
| Surface Water (FWPCA) | 0.05 |
| Irrigation Water (FWPCA) | 5.0 |
| Public Water | 0.05 |

[a] Source: Ref. 109.

[b] Source: Ref. 112.

and surface water is likely to result. Since creosote is highly mobile and persistent, there is increased likelihood of hazardous waste constituents reaching environmental receptors. There is a reported incident of surface water contamination due to improper disposal of creosote wastes, demonstrating that creosote is mobile and persistent and capable of causing substantial harm if improperly managed.

3. It is estimated that 60-115 million lb/year of creosote is contained in the listed wastewater treatment sludge. Thus, substantial amounts of waste constituents are potentially available for environmental release.

### 7.4.1 Process Description

Creosote is produced by the distillation of coal tar, which is produced by the high temperature carbonization of bituminous coal. The two waste streams generated in the production of creosote are: 1) the process wastewater and 2) the sludge resulting from the wastewater treatment plant.

During the distillation process, an appreciable quantity of the water contained in the coal tar (1-2% of the total volume) is boiled off and disposed of along with other process waters. This aqueous waste from the distillation step in the process is the source of hazardous constituents. Creosote wastewater is either discharged to publicly owned treatment works (at smaller facilities) or treated on site in holding ponds (at larger plants). Where on-site treatment is used, ponds are dredged periodically. According to the waste disposal practice prevalent in the chemical industry, these wastewater treatment sludges are transferred to a landfill for final disposal.

On the basis of the estimated annual production of 1,150 million lb/yr of creosote, generation of 1 lb of creosote/12 lb coal tar, and a 1-2%, by volume, water content in coal tar, which is boiled off in the distillation process and disposed of as wastewater, it is estimated that 60-115 million lb of creosote per year are present in the raw process wastewater sent to treatment. Obviously, such large quantities of this waste have the propensity for large-scale environmental harm.

### 7.4.2 Hazardous Components of the Waste

The raw wastewater from the production of creosote is expected to contain varying amounts of creosote, the creosote constituents benz(a)anthracene, benz(b)fluoranthrene and benzo(a)pyrene, all of which are polycylic aromatic hydrocarbons, and other distillation intermediates. The actual composition of the constituents in the wastewater from creosote production depends on the source of the coal used to produce the tar, the design and attendant operating conditions (temperature, coking time, gas collection systems) of the coke ovens, and the design and operating paramters of the still (e.g., the feed rate, temperature, and the blending of various tar distillation fractions). As a result of these factors, the fractional distillation is ordinarily incomplete and a certain amount of creosote residue is present in the raw wastewater.

These waste components could be released from holding ponds into the environment, unless proper management is assured. The migratory potential via ground and surface water, as well as the persistence of creosote have been demonstrated. Creosote and the other waste constituents may thus reach humans and animals in concentrations sufficient to pose a substantial hazard.

The wastewater treatment sludges that remain after biological treatment are also hazardous. The carcinogenic constituents of creosote, namely benz(a)anthracene, benzo(b)fluoranthene and benzo(a)pyrene, are especially likely to be present in the treatment sludge since these constituents adsorb to sediments at very high levels. Where treatment is incomplete, creosote (which is, however, somewhat amenable to biodegradation), is projected to be present in the sludge. If these sludges are placed in a leaking landfill, an unlined holding pond, or an improperly sited facility (e.g., as in an area with permeable soil) the waste constituents may be released.

## 7.5 WOOD PRESERVING INDUSTRY

Wood preserving processes that use creosote or pentachlorophenol as preserving agents generate a wastewater that contains toxic phenolic compounds including penta- and tetrachlorophenol, volatile organic solvents such as benzene and toluene, and polynuclear aromatic (PNA) components of creosote. Treatment of this wastewater results in the generation of a bottom sediment sludge that must be removed for ultimate disposal. The EPA Administrator has determined that wastewater from these wood preserving processes and the resulting bottom sediment sludge are solid wastes that may pose a substantial present or potential hazard to human health or the environment.

The EPA's conclusion is based on the following considerations:[63]

1. The wastewater generated from wood preserving processes using pentachlorophenol as a preservative and the sludge generated from the treatment of this wastewater will contain significant concentrations of toxic phenolic compounds and volatile organic solvents such as benzene.
2. The wastewater from wood preserving processes that use creosote and the sludges generated from the treatment of this wastewater will contain significant concentrations of toxic polynuclear aromatic components of creosote and volatile organics such as toluene. Wastewater and resulting sludges from wood preserving operations that use both creosote and pentachlorophenol as preservatives will generate waste streams that contain all or most of the above contaminants.
3. Polynuclear aromatics, as a group, are known to be toxic, mutagenic, tetratogenic, and carcinogenic. Phenolics are toxic and, in some cases, bioaccumulative and carcinogenic substances. Benzene and toluene are relatively toxic, and benzene is carcinogenic.
4. Approximately 200 million gallons of wastewater are generated annually. About 90% of this wastewater is

treated by treatment methods that generate a bottom sediment sludge.

5. Treatment of wastewater in evaporation ponds or lagoons could lead to the environmental release of hazardous constituents and result in substantial hazards via groundwater or surface water exposure pathways. Evaporation of wastewater in ponds, lagoons, or by other treatment methods, if mismanaged, could lead to the release of hazardous constituents into the atmosphere and result in substantial hazard via an air exposure pathway.
6. Off-site disposal in landfills is the most commonly used disposal method for these sludges. This presents the possibility of the toxic components in the sludge migrating to nearby underground drinking water sources, if the landfill is improperly designed or operated.
7. The EPA has been informed that incineration is another (though less frequently used) disposal method for these sludges. If improperly managed, incineration could result in the release of hazardous vapors to the atmosphere, presenting a substantial hazard.

### 7.5.1 Industry Profile

There are approximately 415 wood preserving plants operated by about 300 companies in the United States. The plants are concentrated in two areas, the Southeast from east Texas to Maryland and along the Northern Pacific coast. These areas correspond to the natural ranges of the southern pine and Douglas fir-western red cedar, respectively.

Approximately 250 million cubic feet of wood are treated each year, principally for railroad ties, utility poles, and lumber for construction materials. Of this amount, it is estimated that approximately 85% is treated with creosote or penthachlorophenol-based preservatives. The total quantity of preservative consumed in 1975 during these treatment cycles exceeded 200 million pounds.

### 7.5.2 Process Description

At plants using creosote or pentachlorophenol-based preservatives, wood products are treated by chemical processes to increase their resistance to natural decay, attack by insects, microorganisms, or fire. Briefly, the treatment consists of debarking, forming, drying, impregnation of preservatives, and storage.

The two major wood preserving processes, producing large quantities of wastewater and sediment sludge, are called steaming and boultonizing. Both of these processes are pressure processes and differ mainly in the way the wood is conditioned before or during the application of the preservative.

Steaming is used principally on southern pines. In this process, the stock is normally steamed for 1 to 16 hours at about 120°C to reduce the wood's moisture content and render it more penetrable to preservatives. After steaming, the preservative is added to the same retort. Condensate removed from the retort after steaming is contaminated with entrained oils, organic compounds, and wood carbohydrates.

In the boulton process, used principally on Western Douglas fir, the wood is already immersed in the preservative, placed under vacuum, and then heated in the retort at approximately 100°C. The vapor removed is composed of water, oils, organic compounds, and carbohydrates from the wood. Contaminated vapors from both the steaming and boultonizing processes are condensed and transported to an oil/water separator to reclaim any free oils and preserving chemicals before treatment and/or disposal of the wastewater, usually an oil/water emulsion.

### 7.5.3 Hazardous Waste Constituents

Bottom sediment sludge from wood preserving plants contains primarily creosote, polynuclear aromatics (creosote compounds), chlorinated phenols, and volatile organic solvents such as toluene and benzene.

EPA has tested samples of bottom sediment sludge and found that it contains polynuclear aromatic hydrocarbons (see below). Many wood processing plants may use both creosote and pentachlorophenol-based processes and thus treat the wastewater generated by these processes in a combined treatment system. Thus, sludge samples from one plant may contain both creosote compounds and phenolic compounds, as shown in the following table:

| Polynuclar Aromatic Hydrocarbons (PAH) | Bottom Sediment Dry Weight (mg/kg), Aerated Lagoon |
|---|---|
| Benz(a)Anthracene | 1250 |
| Chrysene | 9280 |
| Benz(a)pyrene | 5980 |

The following table shows that benzo(a)pyrene may be found in lower concentrations in both oil shale and some coals.[78]

| Oil Shale Materials | (ppb) |
|---|---|
| TOSCO II retorted shale | 13 - 100 |
| GCR retorted shale | 15 |
| Raw shale oil (Colorado) | 30,000 - 40,000 |
| Crude shale oil (TOSCO II) | 3,130 |
| Hydrotreated shale oil (0.25% N) | 6,900 |
| Hydrotreated shale oil (0.05% N) | 690 |

| Coals | ppb |
|---|---|
| High volatile bituminous | 4,200 |
| Low volatile bituminous | 3,150 |
| Lignite | 1,200 |
| Coal tar | $3 \times 10^6$ to $8 \times 10^6$ |

The formation of PAHs in coal conversion processes has been well documented, and several studies have been undertaken for PAH identification and characterization in a number of waste streams at pilot gasification plants.[53] Mass spectroscopic analysis of tars produced in a synthane gasifier have indicated the presence of PAHs. In general, the quantities, chemical structures, and fate of PAHs in coal conversion operations are not well defined and are expected to vary among different plants, depending on the type of coal used, coal conversion process employed, and operating conditions.

Benzo(a)pyrene has been determined to be carcinogenic to experimental animals. It can induce a cancerous skin tumor in mice when 0.25 mg is injected subcutaneously.[109] Benzo(a)pyrene is the most widely accepted indicator of PAH content and biological activity.

## 7.6 PHENOL/ACETONE PRODUCTION INDUSTRY

Distillation bottom tars from the production of phenol/acetone from cumene (heavy tars) contain toxic and potentially carcinogenic organic substances, including phenol and polycyclic aromatic hydrocarbons. EPA's determination of this solid waste as hazardous is based on the following considerations:[109]

1. Approximately 100-200 million pounds of these wastes containing phenol and PAHs from tars are generated annually at 11 plants in the United States.
2. Tars containing PAHs are demonstrated carcinogens and mutagens as well as being toxic. Phenol is a suspected carcinogen and is toxic.
3. There is potential for mismanagement of the waste by leakage during transport or storage, by improper disposal allowing leaching, or by incomplete incinerator combustion.
4. The waste tars persist in the environment, and phenol can spread rapidly in ground or surface water, posing a risk of exposure to these hazardous compounds to humans.

### 7.6.1 Process Description

There are two steps in the manufacturing process: 1) oxidation of cumene to cumene hydroperoxide; and 2) cleavage of the hydroperoxide to form phenol and acetone. Cumene hydroperoxide is the first main reaction product when cumene is oxidized with air at 130 °C in an aqueous sodium carbonate medium. The reaction mix is circulated to a vacuum column where unreacted cumene is separated from the mix and a cumene hydroperoxide concentration of about 80% is obtained in the bottom product. Recovered cumene is recycled to the reactor. Any α-methylstyrene contained in the recovered cumene is separated by distillation and sold or incinerated. However, not all of the α-methylstyrene may be separated at this point. The 80% cumene hydroperoxide cumene mixture is then reacted with 10-25% sulfuric acid at 60 °C and co-mixed with an inert solvent (such as benzene) to extract organic material from the aqueous acid. The mixture is allowed to settle. The acid phase is separated out and recycled to the process. The remaining organic layer is neutralized with dilute sodium hydroxide. The resultant aqueous waste stream containing sodium sulfate, sodium phenate, phenol, acetone, and sodium stearate is separated and sent to wastewater treatment. The crude, neutralized organic layer is then sent to a series of distillation columns where acetone, cumene, phenol, and acetophenone and the solvent are recovered. The first column separates a crude acetone product overhead that is further purified by distillation. The bottoms from the acetone distillation column pass through a waste scrubber to remove residual acetone and inorganic salts. They then pass to a series of columns where the lower boiling hydrocarbons, solvents, cumene, and α-methylstyrene are successively removed, then recovered and sold or recycled and disposed of. The bottoms from the last of the series of columns contain crude phenol, which is refined in the next distillation column from which the purified phenol is removed overhead.

### 7.6.2 Hazardous Properties of the Waste

The bottoms from the phenol still contain phenol, acetophenone, cumyl phenol, phenyl dimethyl carbinol, higher boiling phenolic compounds, and polymers. This mixture may be futher distilled to recover the acetophenone. The still bottoms remaining at the completion of distillation are the waste streams of concern.

The distillation bottoms are a tarry solid in physical form. An EPA study states that these wastes (i.e., the tars generated at the bottom of the acetophenone distillation column) amount to 50-110 g tar/kg of phenol prodcut (100-220 lb tar/ton of phenol).

The waste tars are expected to contain large concentrations of polycyclic aromatic hydrocarbons for the following reasons. Cumene (the essential feedstock material) is itself a polynuclear aromatic hydrocarbon. In the successive steps of hydroperoxidation and acid cleavage, the aromatic ring can open, and other polyaromatic ring structures formed. These are high-boiling substances and will be found in the distillation bottom tars.

## 7.7 SECONDARY LEAD SMELTING INDUSTRY

The emission control dust/sludge from reverberatory furnace smelting of secondary lead products is generated when lead, cadmium, and chromium contaminants found in the source materials are entrained in the furnace fumes during the smelting process and subsequently collected by air pollution control equipment. Dry collection methods generate a dust as a solid residue; wet collection methods generate a sludge as a solid residue. The sludge is usually disposed of on land; the dust is usually recycled for further lead smelting. Before recycling, however, the dust may be leached with acid for zinc recovery, and the resulting waste acid leaching solution containing lead, cadmium, and chromium is disposed of on land. The EPA has determined that these dusts and sludges are solid wastes that may pose a substantial present or potential hazard to human health or the environment. This conclusion is based on the following considerations:[109]

1. The emission control dusts and sludges contain significant concentrations of the toxic heavy metals lead, cadmium, and chromium.
2. Waste leaching solutions from acid leaching of the emission control dusts and sludges contain significant concentrations of lead, cadmium, and chromium, since the acid leaching medium dissolves these heavy metals.
3. The hazardous constituents of these waste streams may migrate from the waste in harmful concentrations, since distilled water extraction procedures performed on samples of the emission control dust and sludge leached significant concentrations of lead, cadmium, and chromium from the dust.
4. The emission control sludge and the waste leaching solutions are typically disposed of in unlined lagoons, thus posing a realistic possibility of migration of lead, cadmium, and chromium to underground drinking water sources. Further, these metals persist in the environment, thereby posing a real danger of long-term contamination.
5. Very large quantities of these emission control dusts and sludges are generated annually (7,151,600 metric tons of sludge and 127,158,000 metric tons of dust in 1977) and are available for disposal as solid waste. There is thus greater likelihood of large-scale contamination of the environment if these wastes are not managed properly.

### 7.7.1 Process Description

Emission control dust and sludge are generated from the manufacture of refined lead, lead oxide, and lead alloy in reverberatory furnaces. In the production process, "soft lead" (low antimony lead) is smelted in a reverberatory furnace from lead residues, scrap lead. In the case of lead alloy, recycled secondary lead emission control dust is a source material. The soft lead is then further processed to either refined lead or lead oxide. In the scrubbing of reverberatory furnace emissions, cadmium, chromium, and lead entrained in the fumes are collected by either wet scrubbing or by baghouse, resulting in a sludge or dust that may be discarded. The EPA attributes the presence of lead, cadmium, and chromium in the waste stream to their presence in the source materials.

### 7.7.2 Hazardous Waste Constituents

The EPA presently does not have data for this industry on the heavy metal concentrations in the waste leaching solution from an operational landfill. Concentrations of these heavy metals in the waste leaching solution, however, can be expected to be significant since the acid leaching medium will readily dissolve heavy metals. Since lead is more soluble in acid than in distilled water, the concentrations of these constituents in the dilute sulfuric acid leaching solution can be expected to be at least as great as, and more likely higher, than concentrations in the distilled water extract. Lead can be bioaccumulated and passed along the food chain but not biomagnified.

Some states are currently operating hazardous waste management programs specifically to regulate cadmium, chromium, and lead compounds as hazardous wastes. Maryland, Minnesota, New Mexico, Oklahoma, and California have final regulations, and Maine, Massachusetts, Vermont, and Louisiana have proposed regulations.

## 7.8 PRIMARY ZINC SMELTING AND REFINING INDUSTRY

The primary zinc industry is composed of plants that employ one of two major zinc manufacturing processes -- electrolytic or pyrometallurgical processing -- to recover zinc metal from ore concentrates. Cadmium and lead contaminants found in the raw materials are carried through numerous processes and are subsequently found in high concentrations in the wastewater treatment sludge generated by the treatment of process wastewater and/or acid plant blowdown, in the electrolytic anode slimes and sludges, and in cadmium plant leach residues (iron oxide). These wastes have been determined to be potentially hazardous to human health or the environment. The EPA's conclusions are based on the following considerations:

1. The wastes contain significant concentrations of the toxic heavy metals cadmium and lead.
2. Cadmium and lead have been shown to leach from samples of these wastes in significant concentrations when the samples were subjected to a distilled water extraction procedure.
3. These wastes are currently stockpiled on site and/or hauled off site to landfills in a manner that could allow the lead and cadmium wastes to leach to groundwater.

### 7.8.1 Process Description

Pyromettallurgical processing entails the following steps: sintering, retorting, refining, and casting. Sintering develops the desired characteristics for pyromettallurgical smelting of the calcine by processing the calcine in a sinter machine where the calcine burns automatically and is fused into hard, permeable sinter. Retorting consists of reducing the calcine in the sinter with carbon in a retort to produce zinc metal. Preheated feed of sinter and coal or coke are fed into the top of the retort; the inside temperature reaches 1300 °C - 1400 °C. Because of zinc's low boiling point (906 °C), it is volatilized as soon as it is formed. In this way, the zinc is purified by separating it from the gangue in the calcine. Zinc from the retort smelting may need further purification for some commercial uses and is purified by distillation in a graphite retort. Molten zinc from the graphite retort is either poured into bars or blocks or is alloyed with other metals and cast.

The sources of hazardous solid waste generated by the pyrometallurgical process are: 1) collection and treatment of acid plant blowdown; and 2) leaching of high-cadmium dusts in the cadmium plant.

In pyrometallurgical plants, roaster off-gas is treated in sulfuric acid plants to control sulfur dioxide emissions. The acid plants produce a salable sulfuric acid bleed stream that must be neutralized. The plant neutralizes the blowdown with lime, which leads to the generation of settled sludge. The sludge contains significant concentrations of cadmium and lead.

The sources of solid waste generated by the electrolytic process are: 1) treatment of preleach residue (this operation occurs at only one plant), 2) the collection and treatment of acid plant blowdown and miscellaneous slurries, 3) the cleaning of the electrolysis cells (anode slimes and sludges), and 4) the filtration of the leach solution.

All zinc concentrations received at zinc plants are roasted to drive off sulfur and convert the zinc sulfide in the concentrate to an impure zinc oxide called calcine. The conversion to calcine in the roaster produces a roaster off-gas stream containing enough sulfur dioxide to permit sulfur recovery as sulfuric acid. The acid production results in a weak acid waste stream from the scrubbing columns that clean the off-gas. This waste is referred to as a bleed stream or acid plant blowdown. The acid plant blowdown is neutralized and thickened, and the solids are recycled or disposed of. Treatment of acid plant blowdown generates sludge that has been designated as hazardous.

All electrolytic plants also generate a waste of anode slimes or sludges from cleaning of the electrolytic cells. Anode slimes/sludges have been designated as hazardous.

### 7.8.2 Hazardous Properties of Waste

Current solid waste control practices are fairly uniform throughout the zinc industry. Of the total solid waste generated, about 90% is controlled through on-site stockpiling, 7% is removed by private and municipal organizations and individuals for various uses (such as winter road sand), and the remaining 3% is hauled and landfilled by private contractors.

Water extractions of the wastes have shown that the wastes could leach potentially hazardous concentrations of toxic metals. This indicates that under the mildest environmental conditions (e.g., neutral pH rainfall) at a mono-disposal site, the wastes may leach contaminants to the groundwater in concentrations that could be harmful to human health and the environment. Where conditions tend to be acidic, the release of heavy metals over the lifetime of a landfill is expected to be even higher than indicated by the water extraction data, since Pb solubilities increase with a decrease in pH. Lead is poisonous in all forms; it is one of the most hazardous of the toxic metals because it accumulates in many organisms, and the deleterious effects are numerous and severe. Lead may enter the human system through inhalation, ingestion, or skin contact. Ingestion of contaminated drinking water is a possible means of exposure to humans as a result of improper management of their wastes.

Table 7.4 compares the levels of lead leaching from coal ash those from solid wastes from the primary zinc smelting and refining industry. These levels appear to be comparable.

Table 7.4 Levels of Lead (mg/L) In Leachate From Coal Ash and Zinc Smelting Processes

| Coal Ash Leachate[a] | | Sludges from Acid Plant Blowdown (Electrolytic)[b] | Sludges from Acid Plant Blowdown (Pyromettalurical Plant)[b] | Anode Slimes and Sludges[b] |
|---|---|---|---|---|
| Spent Solids Residue | Fly Ash | | | |
| 1.1 | 1.6 | 1.0 - 2.1 | 1.3 | 2.0 |

[a] Source: Ref. 112.

[b] Source: Ref. 109.

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