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TREATMENT OF LANDFILL LEACHATES

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Summary

Landfill leachate, which resembles concentrated mixed industrial waste water, threatens national groundwater supplies. Several processes drawn from industrial water treatment have been considered for renovating landfill leachate: biodegradation, chemical and thermal degradation, adsorption, reverse osmosis, and coagulation/precipitation. Since transporting leachate off-site entails considerable risk and expense, these technologies must be evaluated for their applicability to on-site treatment.

Biodegradation has the greatest potential for removing the varied organic compounds found in leachate streams due to its flexibility and relatively wide application in wastewater treatment. Use of combinations of biological processes, such as sequential aerobic and anaerobic treatment, or of biological and physical processes, such as biologically activated carbon, may improve performance. Alternative chemical or thermal destruction processes are more energy intensive and require finer control than biological processes.

Coagulation/precipitation, adsorption, and membrane processes have been studied for treating leachates to remove organic compounds, heavy metals, entrained oil, and colloidal material. Because these processes partition the leachate and, thus, concentrate toxic substances, further treatment of their residues is necessary. They are particularly suited for pretreating influent to a biological process or for polishing a biological effluent.

Much work remains in applying these treatment processes to actual landfill leachates. Design of leachate treatment processes must be tailored to the site and will depend on the quantity and characteristics of treatment residues allowed under the prevailing regulatory environment. The variability of landfill leachates both from site to site and temporally within a site makes leachate treatment a challenging problem.

Introduction

There are thousands of landfills containing hazardous wastes distributed across the country [1]. When water percolates through these landfills, it dissolves or entrains landfill components, such as decaying organic matter, microorganisms, metals, and organic and inorganic compounds [2]. The resulting contaminated water, called leachate, can pollute the underlying groundwater and is considered a major threat to public health [3].

Leachate generation may also occur despite application of landfill isolation techniques such as clay caps, slurry walls, grouting, cementation, or polymer encapsulation. In most cases, isolation methods merely slow the rate of leaching so that leachate collection and treatment are still necessary. Unless contaminated landfill material is physically removed, an expensive undertaking, leachate treatment must play a role in the landfill remediation.

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Landfill leachate is generally a high-strength wastewater containing dissolved and entrained landfill components. Hazardous waste site leachate compositions are presented by Shuckrow [4], Ghassemi [5], and Lamb [6]. They showed that there is substantial variation in leachate composition from site to site and within a site over time. Chian and Dewalle [7] described leachate composition for nine sanitary landfills; leachates were characterized by high concentrations of organic species and heavy metals. They found that the ration of COD/TOC varied from 3.30 for young landfills to 1.16 for older landfills. Development of such parameters for characterizing landfill leachate may prove useful in selecting leachate treatment schemes.

Based on several review articles, five processes are well suited to leachate treatment [2, 4, 9—11]:

Coagulation/precipitation,
Activated carbon adsorption,
Aerobic and anaerobic biodegradation,
Chemical oxidation, and
Reverse osmosis.

Because of the variation in leachate composition from site to site, the remedial process train will generally be tailored to the site and will consist of several unit operations. In this paper, applications of these processes to landfill leachates are discussed. It is important to remember that characterization of leachate plumes through groundwater modeling, analysis of leachate physical and chemical characteristics, and development of leachate recovery systems are all important in selecting a leachate treatment system [8].

Two factors favor the treatment of leachates on-site: the expense of off-site transportation and the reluctance of communities nationwide to permit transportation routes or treatment facilities within their jurisdictions. The desirability of on-site leachate treatment should encourage the development of small scale technology requiring low capital investment. Biological processes are well suited to on-site leachate treatment for removal of organic compounds. The other treatment technologies discussed will find use where the leachate is either untreatable biologically or where pretreatment or posttreatment of a biological stream is required.

Coagulation/precipitation

Extensive literature is available on coagulation as a pretreatment technique [4, 12—16]. Lime, alum and magnesium oxide are commonly used coagulants. Thornton and Blanc [13] performed coagulation studies on leachates from deposits less than two years old containing 50% paper products. Originally these experiments entailed using alum and lime as coagulant and precipitant, respectively, but it was observed that lime possessed a greater ability to reduce suspended solids and color than did alum. Magnesium, calcium, and iron were removed or reduced accompanied by trace amounts of organics. The increase in pH caused by lime addition lead to the forma-

tion of insoluble metal hydroxides and calcium carbonate. The resulting flocs aided in the settling of colloidal material. Thus, more acidic leachates required larger doses of lime. Chian and DeWalle [7] showed that lime coagulation removed trace organic species of molecular weight greater than 50,000 which are found in leachates from newer landfills.

Slater et al. [12] investigated pretreatment of leachate acquired from an industrial landfill in which oil emulsions were noted. Lime was again found to be superior to alum, ferric chloride and magnesium oxide. Lime treatment removed turbidity, dispersed oil, metals, and suspended solids. Slater et al. also examined the use of four polyelectrolytes as coagulant aids to speed up precipitation by increasing the rate of flocculation. Primafloc A-10 (Rhom and Haas) was found to reduce turbidity the fastest. A detention time of ten minutes was demonstrated to reduce turbidity substantially. Following lime coagulation, the leachate pH was approximately 12. Precipitation of calcium from the supernatant and neutralization were accomplished by recarbonating the leachate with carbon dioxide.

Davis [17] used magnesium hydroxide for removing metals from wastewaters characteristic of the metal plating industry. Magnesium hydroxide forms a particulate sludge, rather than a gelatinous precipitate as does lime. Davis points out that this reduces the cost of dewatering the precipitated sludge before ultimate disposal. Other advantages associated with magnesium hydroxide are safety in handling and buffering at pH 9. A disadvantage is that the time for flocculation and precipitation may be substantially longer than that achieved with lime. To remedy this problem, Davis suggests that addition of lime after 20 min can speed up precipitation without increasing sludge bulk.

Sulfide precipitation of heavy metals has been proposed as a replacement for conventional hydroxide precipitation [18, 19]. Advantages include effectiveness over a broad pH range, extremely low solubility of metal sulfides, and reduced detention times. In addition, sulfides, unlike hydroxides, effectively precipitate chelated metals. The presence of sulfide in the effluent and the generation of hydrogen sulfide gas are disadvantages, both of which may be overcome by careful control of the process or by using a combination hydroxide/sulfide precipitation [20].

Bhattacharyya et al. conducted experiments with metal processing wastewater, using sodium sulfide as the precipitant [18]. Precipitation capabilities were increased by addition of lime following the addition of sulfide. Kim has shown that calcium sulfide produces more readily settling precipitates than does sodium sulfide, eliminating the need for lime addition [19]. The calcium sulfide process can be controlled by pH.

Biological degradation of landfill leachate

Biodegradation is the treatment of choice for mineralizing most organic compounds in landfill leachate [5, 21]. Mineralization is carried out by

microorganisms, which can degrade organic compounds to carbon dioxide under aerobic conditions and to a mixture of carbon dioxide and methane under anoxic conditions. Microorganisms are also capable of changing the oxidation state of metals and inorganic compounds and can concentrate heavy metals and hydrophobic compounds through ingestion or adsorption. Microorganisms are ubiquitous, self replicating, adaptable to a variety of leachate compositions, and active at moderate reaction conditions. In addition, biodegradation has a long process history in the treatment of domestic sewage.

Industrial landfill leachate may contain a host of xenobiotics, compounds not normally found in the microbial biosphere [4]. Three factors enable microorganisms to biodegrade many of these foreign organic compounds:
— the flexibility of the microbial genome,
— the diversity of microbial species, and
— the broad specificity of some degradative enzymes.
Utilizing these factors, researchers have demonstrated the microbial degradation of TCDDs, PCBs, and a wide range of pesticides [22—24]. Degradation is not necessarily growth associated [25], as organic compounds may be transformed to microbial storage polysaccharides under nitrogen limiting conditions rather than being mineralized to carbon dioxide. Research in the mechanisms controlling xenobiotic degradation is important in understanding the capabilities and limitations of biological leachate treatment [26].

One of the important elements in xenobiotic biodegradation is the broad specificity of some microbial enzymes, which permits an enzyme catalyzed reaction to occur without providing energy or carbon for cell replication. This phenomenon has been divided into two categories: fortuitous metabolism, in which a growth co-substrate is not obligate, and cometabolism, in which the growth co-substrate is obligate [27, 28]. One of the most thoroughly characterized examples of broad enzyme specificity is the ability of the methane mono-oxygenase enzyme (MMO) to oxygenate hydrocarbons other than methane, its natural substrate. The oxygenated hydrocarbons accumulate stoichiometricaly in the reactor [27]. MMO catalyzed reactions are cometabolic since energy from a co-substrate is required to supply reducing power for the reaction.

Fortuitous or cometabolic biodegradation may account for a significant portion of the removal of xenobiotics in the environment [29]. While numerous examples of cometabolic activity have been described on pure substrates [26, 27, 30], cometabolism has been very difficult to demonstrate in mixed substrate, mixed culture systems since products of the cometabolic reactions of one species may be degraded by another [29]. To encourage cometabolism, easily degradable co-substrates should be included in the leachate prior to biological treatment. Fatty acids, which often occur in landfill leachates, may fulfill this requirement.

If a single microbial species were applied to an industrial landfill leachate, it is unlikely that the microbial enzymatic machinery would be sufficient to degrade all the compounds present [28]. Further, the adaptability of a single

microbial species is limited and the mutational rate is too slow to make single species adaptation practical. In order to increase the diversity of degradative enzymes it is common to employ a mixed microbial population, also known as a microbial consortium or mixed culture. Mixed cultures possess two advantages over pure culture in the degradation of complex substrates. First, the product of an incomplete mineralization by one microbe, such as from a cometabolic transformation, may serve as a substrate for another microbe. Second, the transfer of genetic information between species may enhance the degradative ability of the culture [31]. It has been demonstrated that DDT can be cometabolized to PCPA by one species and that PCPA can be mineralized by another species. A combined culture of the two species results in complete mineralization of DDT [24]. Stable mixed cultures degrading xenobiotics have been isolated in which the microbial consortia degrade a substrate better than the individual species [26, 32, 33].

Many strains have been isolated that can degrade xenobiotics or families of xenobiotics [33]. For example, a white rot fungus studied for its lignin degrading potential has been shown in laboratory studies to mineralize a number of recalcitrant organics, such as a TCDD and DDT [22]. Degradation is carried out by extracellular enzymes whose production is stimulated by nitrogen limitation. Because of the requirements of nitrogen limitation and acidic environment, the fungus is incompatible with many activated sludge derived organisms. Whether such organisms will be useful for degrading mixtures of compounds or will be active in a full scale process has yet to be demonstrated.

Gross genetic changes brought about by the inter-species transfer of genetic material may be important in microbial degradation of xenobiotics. While there are several mechanisms for such transfers, the most important is thought to be conjugation. In this process, loops of extra-chromosomal DNA mediate their own replication from host to recipient microorganisms. Conjugative plasmids, as these DNA loops are known, carry coding for a variety of proteins which, though not required for reproduction, may confer a selective environmental advantage such as heavy metal resistance or extended substrate range [34]. In some cases, non-conjugative plasmids can link to conjugative plasmids and 'piggy-back' from organism to organism [28]. Once a plasmid is transferred, DNA sequences called transposons may play a role in the integration of portions of the plasmid DNA into the genome of the new host. The rapid spread of antibiotic resistance among various classes of microorganisms is an example of the transfer of plasmid-born information.

A number of conjugative plasmids coding for xenobiotic-degrading enzymes have been identified and have been shown to be transferable among microbial species [35]. It has been suggested that plasmid transfer occurs more easily among adsorbed bacteria [26]. Resistances to toxic heavy metals found in microorganisms are often associated with plasmids. These resistances involve either transport (Cd^{2+}, As^{5+}) into the cell or redox reactions (As^{3+}, Hg^{2+}, Cr^{6+}) [36, 37]. Kellogg et al. have presented evidence for the formation

of a novel degradative pathway for a xenobiotic material in a single microorganism through naturally occurring plasmid exchange in a mixed culture system [38]. Such forms of natural genetic engineering are considered more important in the development of new degradative strains than the cloning techniques popularized in medical research [26].

The use of genetic engineering techniques for creation of novel degradative strains has been demonstrated by Reineke et al. [31]. They inserted the toluate degrading plasmid (TOL) or pieces thereof into a strain that could degrade 3,4-chlorocatechol but not chlorobenzoate. The TOL plasmid contained genes coding for the conversion of chlorobenzoate to chlorocatechol, allowing the new strain to completely degrade chlorobenzoate. Two strategies for survivability of engineered genes in mixed cultures are: introduction of a survivable microbe with engineered DNA, and introduction of a transmissable engineered plasmid [26]. The stability and effectiveness of engineered DNA in natural environments has yet to be tested.

The key issues in developing an effective biological landfill leachate treatment process are:

— microbial culture selection and development,
— substrate modification,
— process selection and control.

Due to the complex and undefined nature of industrial landfill leachate, these factors must be evaluated for each site.

The importance of acclimation to leachate biodegradation is emphasized by Schmidt [39] and Grady [28]. Chemical species thought to be biologically recalcitrant may be biodegradable given proper acclimation. Principal mechanisms of acclimation are: macromolecule modification, population selection, and genetic transfer. Modification of cellular components, for example enzyme induction or increased membrane permeability, occurs when a substrate interacts with biological molecules of the cell. The time frame for such interactions is on the order of minutes to hours [40]. Population selection, shifts in the representation of preexisting species, occurs because some species or mutants within a species may be better adapted to a new environment. The time frame depends on growth rates and may range from hours to days for aerobic cultures and from days to weeks for anaerobic cultures [40]. Favorable genetic adaptation, alteration of the microbial DNA, may take periods on the order of months or years [38]. Varma presents a scheme for determining the contributions of induction and population dynamics to acclimation and presents evidence for the dominance of the latter [40].

Lund et al. [41], in acclimation studies on various substituted aromatics, found that acclimation was possible with certain substrates and not with others. However, cultures acclimated on one compound could often degrade a seemingly nonacclimatable compound. One explanation proposed is that some enzymes may have activity on several analogous substrates but may be inducible by only one of those substrates. Thus, to achieve degradation of the non-inducing substrate, the inducing substrate must be supplied.

Grady [28] advocates acclimation by continual seeding from diverse microbial sources under carbon limiting conditions. The objective of continual seeding is to maximize microbial diversity. A microbe which is not a primary degrader of a toxic substrate but can degrade breakdown products may not survive in a culture until primary degraders are well established. As an example of the importance of microbial seed selection, Neyer reports that an activated sludge seed would not survive in a high-salt leachate. Seed obtained from a tanker ballast-water digestor, however, acclimated rapidly [42].

Carbon limiting conditions encourage enzyme induction, place the population under selective pressure for degradation of recalcitrant substrates, and favor simultaneous rather than sequential metabolism of a mixed carbon source [43]. Carbon limiting conditions can be achieved either through continuous culture (chemostat) or through a fed batch reaction.

To facilitate biodegradation, the leachate may require modification, such as pH adjustment, removal or addition of oxygen, amendment with nutrients, or dilution or removal of toxic species. Microbial nutrition is complex and is better understood for aerobes than for anaerobes [44]. Biological processes typically favor a pH near 7. Pretreatment processes to remove inhibitory components include coagulation and precipitation, carbon adsorption, and possibly ozonation [4]. Shuckrow reported that a sewage seed could not be sustained in leachate from the Ott/Story site containing 400—1500 ppm TOC and ppm levels of various chlorinated aliphatic solvents. Substantial TOC removal was achieved biologically when the leachate was pretreated with activated carbon [4].

A variety of biological processes exist to treat leachate [45, 46]. The basic choice is whether to treat a particular leachate aerobically or anaerobically. Both aerobic and anaerobic processes can degrade a wide range of xenobiotics [23, 24]. Aerobic processes are generally superior in mineralizing aromatic compounds; anaerobic processes are superior for short chain aliphatic groups [24]. Aerobic processes have the advantage of speed and ease of control and acclimation. However, aerobic processes accumulate large quantities of microbial sludge that may contain adsorbed organics and heavy metals and may strip volatile compounds. Anaerobic processes produce less sludge and can provide energy through methane production. They also reduce sulfate to sulfide which is a powerful precipitator of heavy metals. However, because of their low reproduction rates, anaerobes require a long start-up time and are sensitive to toxic shocks [44, 47]. Both aerobic and anaerobic processes have been shown capable of degrading portions of industrial landfill leachate, but neither has seen extensive field use [1, 48].

The rate of mineralization of organic carbon in a biological process depends on the concentration of active cell mass. The maximum cell mass sustainable in a process will depend on nutrient availability, gas transfer, and toxicity of the leachate. In aerobic and anaerobic treatment lagoons, no provision is made for concentrating the suspended cells. Therefore, lagoons must be large to effect organic removal. The advantage of lagoons is that

little maintenance or control need be exercised other than periodic dredging of the microbial sludge [4].

Cell concentration can be increased either by cell recycle or by biological film processes. Cell suspension with recycle has been employed in the traditional activated sludge process and in the upflow anaerobic reactor (UAR) both of which recycle the cell mass using the natural flocculation properties of cells. Activated sludge processes on 92 industrial waste streams have shown an average TOC removal of 86% [49]. Leachate treatability tests presented by Goltz et al. showed substantial removals of several organic components from complex landfill leachate by an aerobic activated sludge reactor [50]. Biological treatment has also been applied to leachate from Love Canal [4]. Chiesa et al. have shown that carbon limited environments encourage the growth of nonfilamentous microorganisms, which settle faster than filamentous microorganisms [51]. Control of cell settling in biodegradation is an important area of study.

Biological film processes, including rotating biological contactors, trickle beds, and anaerobic fixed film reactors, are another means of increasing cell mass in a continuous flow system. An advantage of biological film processes is the long biomass retention times achieved without the need for a settling tank. Bouwer et al. have demonstrated the use of an anaerobic fixed film reactor to degrade halogenated aliphatic compounds such as 1,1,1-trichloroethane [52]. Kaschak et al. report that a comparison of activated sludge, a rotating biological contactor, and an aerated lagoon for removal of phenols and organic extractables showed comparable removal efficiencies. Activated sludge was preferred on the basis of economics [53].

The reduction in organic carbon achievable by microorganisms is limited by the minimum concentration required to maintain cellular metabolic processes [54]. While microbial species known as oligotrophs can operate at low substrate concentrations, they may not be capable of reducing contaminant concentrations below water quality standards. Two methods have been proposed to circumvent the biological maintenance barrier to leachate degradation. In the first, activated carbon is added to a biodegradation reaction [55, 56]. There are three potentially beneficial effects of adsorbent addition: organic carbon is concentrated for microbial attack in the microenvironment around the adsorbent particle, the concentration of potentially inhibitory organic compounds in the bulk solution is lowered, and the carbon particles serve as a surface for microbial growth [57]. Dienemann et al. used a mixed aerobic/anaerobic system in a column of soil and 5% activated carbon to treat both an industrial landfill leachate and an alkaline extract of the landfill sludge [58]. Removals of 90% to 95% of the organic carbon was achieved in both cases. A second method, the unsteady state fixed film reactor, has also been shown capable of reducing effluent concentrations below maintenance requirements [59].

Leachates can also be degraded biologically in-situ. Conditions within the landfill are controlled to encourage microbial activity and leachate is recircu-

lated thru the landfill. Recirculated leachate may require nutrient amendment, neutralization, or heavy metal removal. Aerobic microbial activity occurs at the landfill surface while anaerobic activity occurs in the landfill interior. Kosson et al. showed that while aerobic and anaerobic activity individually only degraded 60—90% of a leachate, a combined aerobic/anaerobic soil filter was capable of degrading 95% of the leachate [63]. Recirculation combined with anaerobic activity may stabilize heavy metals through precipitation of heavy metal sulfides [60]. Because aerobic biodegradation is faster and better understood, methods for encouraging aerobic activity within a landfill by addition of hydrogen peroxide or air microbubbles have been investigated [61]. Subsurface aeration wells have also been employed to encourage in-situ degradation. No evidence for their success was presented [62].

Biodegradation is a promising method for primary removal of organic compounds from landfill leachate. However, some organic compounds are resistant to biological attack. In addition, biological sludge resulting from biological processes may become a disposal problem, particularly because of its capacity to store adsorbed undegraded hydrophobic organic species and heavy metals. No biological leachate treatment processes yet take advantage of microbial transformations or adsorption of heavy metals though suitable microorganisms have been studied in the laboratory [64]. Biodegradation processes are still relatively unsophisticated and potential exists for combining various types of microbial process schemes for selective component removal.

Adsorption

Carbon adsorption is the most extensively used physical—chemical means for removal of organic constituents from wastewater and leachate [16]. Adsorptive capacity of the carbon depends on preparation, storage conditions, pore size, and surface area [65]. For solutes, adsorptivity increases as the solubility decreases. Thus, for hydrocarbons, adsorption increases with molecular weight. Solution pH also influences adsorptivity, possibly to the extent that it affects solubility. O'Brien et al. [66] and Shuckrow et al. [16] present extensive data describing adsorption of organic compounds on granular activated carbon (GAC), including two case studies describing the successful removal of trichloroethylene, cis-1,2-dichloroethylene, and tetrachloroethylene by activated carbon [66].

Adsorption of inorganic species has also been investigated by Netzer and Hughes [67], who evaluated ten commercially available activated carbons for removal of lead, copper, and cobalt from aqueous solutions. Barney and Cheney NL1266 was superior. Adsorption of these metals was determined to be sensitive to pH, with maximum removal at pH 4 or higher. Copper hindered the adsorption of cobalt and lead. Treatment techniques previously mentioned (coagulation/precipitation) are more commonly used for metal removal.

Gorgol and Ahlert investigated the applicability of GAC adsorption to leachate treatment [68]. To eliminate pore blockage due to leachate turbidity and entrained oil, pretreatment by coagulation, recarbonation, and pH adjustment was performed with lime, carbon dioxide, and sulfuric acid, in sequence. By deaerating the adsorption columns with either water or the liquid to be treated, channeling, high pressure drops, and permeate breakthrough of adsorbate was avoided or reduced.

Randtke and Jepsen [69] demonstrated that alum coagulation increases the adsorptive capacity of GAC for fulvic acids, which are a major constituent of leachate [70]. Coagulation, with little or no reduction in TOC, shifted adsorption isotherms positively. The authors hypothesized that this phenomenon was due to organo—aluminium interactions.

Carbon regeneration may improve the economics of activated carbon treatments. Smithson describes several regeneration techniques [71]. Of these, thermal regeneration, in which adsorbed organics are volitilized or oxidized at high temperature, is the most common [72]. The cost of carbon attrition and energy consumption for thermal regeneration have encouraged development of other regeneration techniques. Martin and Ng [72] studied organic and inorganic chemical regeneration on activated carbon exhausted with mono-substituted benzene compounds. They found that organic solvents such as carboxylic acids, were the most effective regenerants. They also found that desorptive partitioning increased with decreasing molecular weight which they attributed to penetration into the smaller carbon pores. Desorption of pesticides from activated carbon with supercritical carbon dioxide has been reported, with a recovery of 70% of the adsorptive capacity of the virgin carbon [73]. Decompression of the carbon dioxide is all that is required for its reuse. The disadvantage of supercritical regeneration is the high operating pressure required.

Adsorption on synthetic resins has been used for leachate and wastewater treatment. TOC removals in excess of 90% have been demonstrated using three types of resins in series, each with a different adsorption function [74]. Polyacrylamide resins commonly adsorb low molecular weight organic acids while synthetic carbonaceous adsorbents remove chlorinated organics, such as trichloroethylene, 1,1-dichloroethane, and cis-1,2-dichloroethylene [75].

Membrane processes

Reverse osmosis (RO) has been used in the treatment of wastewater to remove salts [76]. Chian and DeWalle consider it the most effective physical—chemical means for treatment of leachate from sanitary landfills and industrial landfills [7]. Slater et al. [76] performed reverse osmosis experiments on industrial landfill leachate using tubular cellulose acetate membranes. Pretreatment by lime coagulation and recarbonation, as described earlier, was executed prior to reverse osmosis [12]. Total recycle was demonstrated to reduce permeate ion concentration by 95%, dissolved solids by 98%, chemical

oxygen demand by 68%, and total organic carbon by 59%. No fouling was observed throughout the study. A second experiment was performed with a membrane of lower rejection. The leachate under observation had higher concentrations of organic and inorganic species. Dissolved solids rejection was 88%, but COD and TOC rejections were nearly the same as in the previous results.

Slater et al. also studied RO without recycle to achieve 75% recovery. Leachate TDS and conductivity were lower than for the two previous experiments. A membrane similar to that in the first experiment was used. Both TDS and conductivity of the permeate rose as the leachate feed became more concentrated. If was suggested that an anti-scalant be used to obtain recoveries exceeding 75%.

Fang and Chian conducted studies in which membranes of different compositions were evaluated for concentrating various aqueous polar organic compounds [77]. Sulfonated polyphenylene oxide and poly-2,2-(m-phenylene)-5,5-bibenzyimidazole membranes performed poorly. Cellulose acetate in tubular and flat configurations and cellulose acetate butyrate and cellulose triacetate exhibited poor separation capabilities. Aromatic polyamide membranes yielded approximately 50% separation. Cross-linked polyethylenimines were superior to the other types studied, yielding 70—80% overall separation.

High pressure must be maintained to effect reverse osmosis, resulting in high operating costs. Siler and Bhattacharyya [78] have studied thin-film composite membranes which require much lower pressures than cellulose acetate membranes. Separation capabilities are comparable.

Syzdek and Ahlert investigated the feasibility of ultrafiltration as pretreatment for reverse osmosis [79]. The leachate used for this study was from an industrial landfill. Membranes of polyelectrolyte, polyolefin, polyaromatic, and cellulose acetate were evaluated for their ability to withstand exposure to the leachate. Only the polyelectrolyte membrane experienced a retention decrease after exposure to the leachate. Filtration of the leachate through membranes of decreasing molecular weight cutoffs ranging from 300,000 to 2,000 was performed in series. Fouling by gel formation at the surface of the first three membranes of largest molecular weight cutoff was observed. The gel composition was primarily inorganic indicating less than complete removal by chemical pretreatment. To reduce fouling, it was recommended that ultrafiltration be performed in the presence of stirring or by increasing the flow rate in a tubular UF set up. With leachate attenuated with distilled water, pressure can be increased without fouling, resulting in greater flux. Temperature was determined to have little effect on gel formation.

Maier and Cussler suggested employing cross-linked polyacrylamide gels to concentrate wastewater streams [80]. When the gel absorbs water from solution it expands, prohibiting the passage of macromolecules. It is regenerated by lowering the pH, resulting in gel contraction and release of the absorbed water. Neutralization of the gel prepares it for reuse. Solute rejection can be controlled by varying the degree of cross-linking. At the time of publication,

the application of reversible gel absorption to wastewater constituents had not yet been attempted.

Chemical treatment of landfill leachate

The following chemical conversion techniques have been examined for treating landfill leachate [10]:

wet oxidation,
ozonation,
hydrogen peroxide treatment, and
chemical reduction.

Typically, these processes are energy intensive or require special handling. They may find application to very high strength or biologically recalcitrant leachates [4, 8, 10].

Wet air oxidation involves aqueous phase oxidation, at 175°C to 320°C and 20 to 200 atm pressure [4]. Typical residence times are one to two hours. Besides maintaining the waste in the liquid phase, the pressure increases the concentration of the oxidant, dissolved oxygen, which is supplied as high pressure air or pure oxygen. The influent waste stream must contain 15% chemical oxygen demand to be self sustaining; otherwise heat must be supplied to the process [81]. A catalyst, such as copper, may be included [10]. Oxidation products include carbon dioxide, alcohols, ketones, aldehydes, and carboxylic acids. Examples of wet air oxidation have been presented by Rappe [82], Dietrich [83], and Canney [81]. Dietrich lists compounds oxidizable by wet air oxidation. Waste streams tested include coke plant wastewater and pesticide production wastewater. The effluent from the process may require additional biological treatment. For example, effluent from the oxidation of a 110,000 ppm COD pesticide waste stream had a residual COD of over 5000 ppm.

Ozone, generated by electric discharge in air, has also been used for the oxidation of organic solutes. Rice discusses the chemistry of the ozonation of organic compounds and lists classes of compounds resistant to oxidation by ozone, such as halogenated aliphatic solvents [84]. Ozonation has been evaluated for removal of cyanide and phenolics from waste streams [85, 86]. Due to solubility and mass transfer limitations and generation costs, ozone is only applicable to waste streams with less than 1% oxidizable materials [4]. Ozone is toxic to humans and corrosive; thus, special equipment and precautions must be taken in its use. Ozonation of some substrates, such as certain pesticides, can yield hazardous products. The oxidizing action of ozone can be increased significantly if it is used in conjunction with ultraviolet radiation or hydrogen peroxide.

Hydrogen peroxide is another agent for oxidizing organic materials. Like ozonation, peroxide is best suited to treating dilute streams and must be handled carefully. The use of hydrogen peroxide for the destruction of phenolics has been reported [87]. Both hydrogen peroxide and ozone are

preferable to chlorination for oxidation of leachates, since no chlorinated byproducts are produced.

Reduction of organics can be accomplished by activated catalytic metals such as aluminum, zinc, and iron at room temperature [88]. Catalytic reduction has been used to dehalogenate pesticides, such as DDT, in dilute (<1 ppm) solutions. Reaction products depend on the reductant. Other substrates for catalytic reduction include halomethanes, PCBs, chlordane, kepone, atrazine, dimethylnitrosamine, and dinitrophenols [88]. Removals to 1 ppb have been observed. A typical process consists of neutralization followed by passage of the waste stream through a reductant bed with retention time of the order of 5 min. The optimal pH is about 7. At low influent organic concentrations, dissolved metal catalyst in the effluent is on the order of 1—5 g/l. Thus, a metal precipitation step is required subsequent to reduction. Reduction may be a useful step for treating refractory materials prior to biological treatment or for reducing heavy metals to less toxic oxidation states. Sulfer dioxide converts chromium(VI) to chromium(III) which can be precipitated by chromium hydroxide by addition of sodium hydroxide [10].

Leachate renovation processes

Of twenty potential leachate treatment unit operations listed by Shuckrow, only six were thought to be practicable: biological treatment, chemical coagulation, carbon adsorption, membrane processes, resin adsorption, and stripping [4]. Both Shuckrow et al. [4] and Chian et al. [21] recommend biodegradation as the most flexible and economical leachate treatment. In practice, however, carbon adsorption has been the principal leachate renovation process. Of 23 case studies cited in a recent EPA report, 16 involved removal or isolation of the landfill material without leachate or groundwater treatment. Of the remaining cases, three employed activated carbon, one combined activated carbon and activated sludge, and three utilized local POTWs for groundwater treatment [62].

Carbon adsorption treatment was used at the Love Canal and Goose Farm sites. At Goose Farm in New Jersey, a process consisting of flow equalization, neutralization, activated carbon, and air stripping was used to treat leachate resulting from landfilled rocket propellants and specialty chemicals [62]. The process removed 62% of the influent TOC. Spent carbon was disposed off-site.

McDougall described the treatment process used at Love Canal which consisted of neutralization, clarification, multimedia filtration, and carbon adsorption [89]. Each adsorber contained 20,000 lb (9,090 kg) of carbon and could treat 662 l/min. The influent TOC of 700—900 ppm was reduced to 50—100 ppm in the effluent. Carbon use averaged 87 lb/h (39 kg/h). In Lathrop, CA, a carbon adsorption process was used to treat dibromochloropropane after UV destruction was rejected due to low performance and scaling. Reduction of the DBCP to 4—6 ppb required 5,000—11,000 pounds (2,270—4,990 kg) of carbon per month [62].

Several process trains involving biological treatment have been studied at pilot scale. An activated sludge process was used to treat groundwater contaminated with isopropyl alcohol, methylene chloride, acetone, butanol, and dimethyl analine [62]. Contaminated air from the activated sludge unit was sent to a carbon adsorption column. Effluent from the activated sludge process was injected into the ground to encourage in situ degradation of contaminants and aeration wells were operated to supply oxygen. No information on the effectiveness of the in situ treatment has been reported.

Neyer reported on the treatment of a brine groundwater [42]. Effluent from activated sludge was polished using a fixed aerobic filter followed by a mixed media filter and carbon adsorption. TOC removal was reported at 85%. Keenan et al. describe a process to treat sanitary landfill leachate consisting of equalization, precipitation, air stripping, neutralization, activated sludge, and chlorination [15]. Sanitary landfill leachate with an influent COD of 18,488 mg/l was treated at a maximum rate of 20,000 grams per day. Greater than 95% of BOD, COD, and suspended solids were removed and 88% of the influent Kjeldahl nitrogen. The major problem was scum removal from the activated sludge process. The use of cotreatment of landfill leachate in a municipal wastewater plant was investigated by Schuk and James [90]. They used a synthetic leachate which they found to be treatable as long as the proper nutrients were added and the oxygen transfer capacity of the aeration basin was not exceeded. Addition of leachate resulted in a significantly increased mixed liquor respiration rate.

Argo describes a process for treating the effluent of an activated sludge process consisting of lime clarification, air stripping, neutralization by recarbonation, chlorination, mixed media filtration, and a partial reverse osmosis or activated carbon. Effluent TOC concentrations could be reduced to 5 ppm [14].

Shuckrow et al. evaluated the use of chemical coagulation, aerobic and anaerobic biological treatment, carbon adsorption, resin adsorption, stripping and ozonation applied to the Ott/Story leachate [4]. The leachate contained a variety of volatile organic species, and acid and base extractables. A process train of activated carbon followed by aerobic biological treatment was feasible. The combined process resulted in better performance than either unit process individually [4].

Perhaps because of the legal questions and uncertain regulatory climate surrounding chemical and waste dump closures, field experience with existing leachate renovation technology is scant. However, as the public awareness and concern grows, economic forces accruing from statutory penalties may remedy this situation.

Conclusion

Biological and chemical oxidation, adsorption, coagulation, and membrane processes are discussed with regard to leachate treatment. Because of the

complexity and site-to-site variation of leachate, the treatment process must be tailored to the site. On-site treatment is favored due to the expense of transporting leachates off-site.

Of the treatment processes discussed, biological leachate treatment is generally the most cost effective means for destruction of organic leachate constituents. Aerobic treatment is easier to control and has a longer industrial waste treatment history, but anaerobic processes, because of low energy consumption and minimal sludge production, deserve consideration. An understanding of microbial interaction and selection is important in developing and controlling mixed microbial cultures. The role of genetically engineered organisms is uncertain given their instability and the complexity and variability of leachate composition. Alternative chemical or thermal leachate oxidation techniques may be suitable for certain types of leachates but operate at more extreme conditions and thus require more stringent control and safety precautions.

Coagulation/precipitation schemes have been demonstrated for removing both heavy metals, colloidal material, and entrained oil. Lime is the most commonly used coagulant. Settling of calcium flocs can be increased through addition of polyelectrolytes. Calcium or sodium sulfide gives more complete precipitation of heavy metals than lime though lime addition speeds settling of heavy metal sulfides. Carbon adsorption and membrane processes are also capable of removing a variety of organic and inorganic leachate constituents. However, coagulation, adsorption, and membrane processes do not destroy the partitioned leachate constituents so that further processing or disposal is necessary.

While none of these processes is adequate by itself, combinations have been shown capable of treating a variety of leachates. An important consideration in process selection is the nature and quantity of residuals. Further developments in leachate treatment processes will be strongly influenced by the regulatory requirements surrounding hazardous and chemical waste dumps.

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