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EVALUATION OF CEMENT, LIME, AND ASPHALT AMENDED MUNICIPAL SOLID WASTE INCINERATOR RESIDUES

1.1.1.

BY

PETER J. HOLLAND B.S.C.E. United States Air Force Academy, 1983

THESIS

Submitted to the University of New Hampshire in Partial Fulfillment of the Requirements for the Degree of

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in

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30 JUNE 1989 Date

DEDICATION

How do I begin to thank the one person who has walked with me every step of the way. Through the fourteen month separation that was Korea, to the trials and tribulations of this accelerated Master's program. You encouraged me when I was frustrated, you motivated me when I lacked ambition, and you inspired me with your undying love. My wife, my best friend, Jenny.

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ABSTRACT

EVALUATION OF CEMENT, LIME, AND ASPHALT AMENDED MUNICIPAL SOLID WASTE INCINERATOR RESIDUES

by

Peter J. Holland University of New Hampshire, September, 1989

The ability of type II portland cement, hydrated lime, and AC-10 asphalt to solidify/ stabilize municipal solid waste (MSW) incinerator residues was evaluated. Column leach tests were performed on unamended combined residue (92% bottom to 8% fly ash by volume), as well as crushed 17% cement and 5% lime amended residues. /The columns were run in a downflow, fully saturated configuration using a pH 4.0 synthetic acid rain leaching media. ⁴Leachate withdrawal was controlled at 0.13 ml/min until final liquid/ solid ratios of 0.6, 0.4, and 0.9 was achieved in the cement, lime, and residue control columns, respectively. The leachates were analyzed for metal (Pb, Ni, Zn, AL, Ca, Mn, Na, Cd, Cu, Fe) and anion (CL, SO_{4}^{T} , NO_{3}^{T} , PO_{4}^{T} , Br) concentrations, alkalinity, pH, redox potential, and conductivity. Successful immobilization of all tested metals except lead, aluminum, and nickel, were seen in the cement amended leachates, while only zinc and manganese were leached from the lime amended columns in concentrations exceeding those found in the Conte residue control.

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Finely ground (> 0.3mm) 33%, 17%, and 11% cement amended residues; 10%, and 5% lime amended residues; and 26%, 22%, and 18% asphalt amended residues were subjected to batch leaching tests to compare relative leachable metal fractions and kinetic release profiles under controlled pH conditions. The test results revealed little differences in metal mobility between wastes of varying percent amendment addition. In general, cement, lime, and asphalt amended residues demonstrated reduced mobility for cadmium, manganese, and aluminum relative to the concentrations measured in the control leachates.

Contri

Compressive strength tests were performed on replicate specimens of all cement, lime, and asphalt amended matrices as a measure of residue solidification. Cement and lime amended specimens exhibited average 28 day compressive the Ses. And strengths ranging from 280 to 780 psi, and 4 to 25 psi, respectively. Increasing compressive strengths were realized commensurate to the percent cement and lime amendment. Asphalt amended specimens demonstrated similar increasing 28 day compressive strength relationships to a maximum at 23% asphalt content. At this asphalt content, 28 day compressive strengths of 580 psi and Marshal stability results of 2000 pounds (flow=23) were achieved.

Additional MSW incinerator residue solidification/ stabilization data is essential to the development of proper management strategies capable of combating the potential solid waste crisis of the future. Further refinement of leaching methodology should provide insights into metal release mechanisms associated with solidified/ stabilized MSW incinerator residues and enhance understanding of long term leaching behaviors.

INTRODUCTION

Management and disposal of municipal solid wastes (MSW) is fast becoming one of the key environmental challenges facing many state and local governments today. Increasing MSW production coupled with decreasing resources with which to handle the wastes, have prompted many communities to seek alternative management strategies. Two processes which figure prominently in many future MSW management strategies are recycling and incineration.

The concept of recycling has often been overlooked as a viable means of reducing the volume of MSW produced in the United States. EPA (1987) estimates that of the 150 million tons of MSW produced in 1984, only 10% was recycled. Many communities are placing greater emphasis on source reduction/ recycling programs in light of the projected increases in MSW which are expected to exceed 290 million tons annually by the turn of the century (EPA, 1987).

The potential for far greater MSW volume reduction may be realized through the use of incineration. Hjelmar (1987) demonstrated that incineration may effectively reduce the volume and weight of MSW by 90% and 70%, respectively. Additionally, the combustion of MSW represents a potential source of steam and heat generation.

The benefits of MSW incineration are not accrued without environmental costs. Release of toxic constituents to the environment due to the combustion of MSW may occur either directly, through stack emissions, or indirectly through contaminant leaching of the resulting residues under a landfill scenario. The technology exists to effectively eliminate many toxic air pollutants from the stack gases, while the control of contaminant leaching from MSW incinerator residues remains the subject of significant public and scientific concern. Much of the emphasis has been focused on solidifying and/or stabilizing the incinerator residues, which are more concentrated with potentially toxic metals than their raw MSW predecessor.

Some available solidification/ stabilization processes include the use of thermoplastic encapsulation (asphalt), glassification, or cement-based and lime-based fixation (EPA, 1979). Each of the amendments possess the potential to physically solidify MSW incinerator ash and chemically stabilize the metals as insoluble mineral phases within the waste matrix.

The ability of cement, lime, and asphalt to impede the release of metals from their respective residue matrices depends largely on the physical and chemical properties of the binding mechanism, as well as the matrices' environmental conditions. If the amended waste does not contain sufficient pozzolans for lime solidification, or if organics in the cement residue mixes are excessive, a low

strength material will result due to the inability to form complex calcium and aluminum silicates within the matrices. The weaker the matrix, the greater susceptibility to environmental attack and hence metal mobility (Cote, et al, 1986).

Similarly, leaching of metals from asphalt amended MSW incinerator residues may occur due to insufficient coverage of the residue by the asphalt binder. This defeats the asphalt's primary metal immobilization mechanism, microencapsulation, and permits direct contact between the residue particles and the environment (Cullinane & Jones, 1989).

Several researchers have identified the primary metal release mechanisms from solidified/ stabilized residues as outward diffusion from the pore water, and to a lesser extent, dissolution of metals from the matrix surfaces (van der Sloot et al., 1987; Cote, 1986; Cullinane & Jones, 1989). Important factors affecting those release mechanisms include the stabilized material's alkalinity, pH, and redox conditions, as well as the open porosity, and surface to volume ratio. In a landfill or reuse scenario, these properties are largely affected by the conditions and characteristics of the solidified matrix's chemical environment and hydraulic regime. Therefore, selection of a solidification/ stabilization process for MSW incinerator residues must consider the resulting matrix's chemical and

physical properties and the site specific environmental conditions.

The objective of this research was to compare various mixes of cement, lime, and asphalt amended incinerator residues with regard to compressive strength and metal leachability. The former was determined using conventional cylinder tests, while the assessment of metal leachability was based on both column and batch leaching experiments.

Solidification of the combined residue with lime and cement produced a material with a compressive strength commensurate to the percent amendment. Asphalt amended specimens demonstrated increasing compressive strengths and Marshall stabilities with increasing percent addition to a maximum at 23% asphalt content.

Column leaching studies of the crushed amended materials suggest that cement amending will attenuate certain metal constituents (Na, Mn, Ca) of the incinerator residue, while promoting the mobilization of other metals (Pb, Al) when contacted by a mildly acidic leaching media. Under similar conditions, the lime amended residues demonstrated reduced manganese and sodium mobility, while maintaining leaching patterns characteristic of the unamended residue for all other tested metals.

Batch leaching of finely ground (> 0.3mm) cement, lime, and asphalt residues revealed little differences in

metal mobility between wastes of varying percent amendment addition. In general, cement, lime, and asphalt amended residues demonstrated reduced mobility for cadmium, manganese, and aluminum relative to the concentrations measured in the control leachates.

The further refinement of metal release mechanisms associated with solidified/ stabilized MSW incinerator residues is required to enhance understanding of long term leaching behaviors. This data is essential to the development of proper MSW management strategies capable of combating the solid waste crisis.

CHAPTER I

LITERATURE REVIEW

MSW Handling & Disposal Practices

Direct landfilling of municipal solid wastes (MSW) represents society's traditional approach to MSW management. Favorable economics, regulatory policies, and sheer convenience has been the impetus behind such handling and disposal practices (Wiles, 1987) . Past scientific studies have linked numerous incidence of adverse human health to the continued pursuit and operation of traditional MSW landfills (Ehrig, 1983; Jackson & Lynch, 1982). Recent legislation has provided incentives and controls to stem the use of landfilling as the sole means of MSW management while encouraging treatment processes that reduce MSW volume prior to land disposal.

Source reduction, recycling and incineration of the raw MSW represent viable volume reduction processes. The past use of these technologies for managing MSW can be best characterized as limited. As of 1984, the U.S. Environmental Protection Agency (EPA) reported less than 10% of all MSW produced annually were recycled, while only 5% were incinerated (EPA, 1987). These percentages appear to be increasing primarily through heighten public awareness of MSW related issues.

The media is largely responsible for the present increase in public education concerning MSW management. News stories of environmental interest groups lobbying for reductions in the use of plastic packaging, and local communities instituting mandatory recycling programs are testaments to the public's willingness to participate in MSW volume reduction processes. It is estimated that 30 - 50% MSW volume reduction is achievable through increased government and public participation in source reduction and recycling programs (Dennison, 1988). Unfortunately, effective volume reduction and recycling of MSW will not completely erase the potential solid waste crisis we face in the future. That is why many municipalities are turning to incineration as a means of reducing the quantity of MSW ultimately requiring disposal.

EPA estimates over 50,000 tons per Gay (tpd) of MSW were incinerated at 111 facilities in the U.S. during 1984 (EPA, 1987). Plans to construct an additional 210 facilities by the year 2000, will increase processing capacity to nearly 260,000 tpd, and bring the total number of operational MSW incinerators to over 300. Unfortunately, this projected growth in the MSW incineration industry will still be insufficient in meeting the estimated increases in MSW. Even if a 50% reduction of MSW is realized through source reduction/ recycling, an estimated 30 million tons of MSW will be left unprocessed (EPA, 1987).

Environmental Issues Concerning MSW Incineration

Municipal solid waste incineration is not a panacea, nor does it face overwhelming acceptance by the public. Major environmental concerns are primarily focused on air and residue emissions resulting from the combustion process.

Air Quality

As with any high temperature combustion process, the release of harmful pollutants into the atmosphere is an ever present possibility. Savage et al. (1988) lists the principal air contaminants of concern in MSW incineration as: carbon dioxide (CO_2) , carbon monoxide (CO), acid gases, oxides of nitrogen (NO_x) and sulfur (SO_x) , particulates, and toxic organic compounds. Toxic organic compounds are of particular concern due to their strong carcinogenic effects at relatively low levels (Waldbott, 1973; EPA, 1986). Recent studies cite several chlorinated aromatics, specifically polychlorinated dibenzodioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and polychlorinated biphenyl (PCBs), as the principal toxic organics associated with MSW incinerator emissions (Commoner et al., 1987).

Various control technologies have evolved that sufficiently remove these contaminants from the MSW combustor stack emissions in accordance with regulatory

standards (Wark & Warner, 1976). Current legislation requires the incorporation of appropriate control devices in the design of new state of the art facilities. Facilities already in operation are being reviewed on an individual basis and required to retrofit control measures where appropriate to adhere to federal/local restrictions (EPA, 1987b).

Residue Emissions

The combustion of MSW yields two distinctive waste streams that differ in grain size, morphology, and composition. The bottom ash fraction typically comprises 90 - 99% of the total residue produced by mass burn facilities, with an effective particle diameter of 2 mm, and a predominately plate-like morphology (Eighmy et al., 1988). Flakes appear to be the principal structure of the fly ash particles, maintaining an average effective diameter of 0.08 mm.

Ultimate disposal of the residues is usually accomplished by landfilling. Serious concerns over the ecological impact of such practices have arisen due to the potential of surface and groundwater contamination from release of the toxic constituents commonly associated with the residues.

These concerns are amplified given the hazardous classification typically assigned to fly ashes on the basis

of lead and cadmium toxicity (EPA, 1987). Fortunately, the largest residue fraction, bottom ash, is more benign and characteristically does not carry the hazardous label. Combined ash streams of 1 to 10 % fly ash to 99 - 90% bottom ash mixtures assume a hybrid characteristics, earning hazardous classification based on overall fly ash percentage (Thompson, 1989).

The two major environmental issues confronting MSW incinerator residues are not mutually exclusive. The overall treatment and quality of the stack emissions has a direct bearing on resulting residue's leachability. Certain acid gas treatments common to emission control technologies, often promote metal mobility from the landfill MSW incinerator residues. Additionally, the levels of NO_x and SO_x emissions may indirectly impact metal leachability through the formation of acid rain which infiltrates landfilled residues and increases metal solubility in the resulting leachates.

Incinerator Siting

A more intrinsic, yet very real problem exists concerning MSW incineration based on geography. EPA (1987) estimates that the siting of MSW incinerators based on need/ MSW generation would require operating 66% of the facilities in the north eastern states of New Jersey, Pennsylvania, and the New Englands. Localization of facilities in these

highly industrialized and developed areas may potentially aggravate the regional air quality and adversely impact the already shrinking real estate market. Inevitably, solving these problems will require a tremendous financial and political effort. Through the continued refinement of available air and residue control technology and the strengthening of public awareness and support, effective MSW management strategies can and will evolve.

Ash Composition

Metal Partitioning

The distribution of constituents among the fly ash, bottom ash, and flue gases were studied by Brunner & Monch (1986) and the NUS Corporation (1987). In both studies, the partitioning of constituents was found to depend on the volatility of the constituent, incineration conditions (e.g. combustion temperature, burn time, etc) and the removal technology employed by each facility. A summary of constituent partitioning determined in the Brunner & Monch (1986) study is shown in Figure 1.1.

Of the constituents measured, only iron and copper remained primarily associated with the bottom ash. Chloride, carbon, and mercury appear predominately in the stack gases, while cadmium demonstrates strong partitioning to the fly ash. Fluoride and sulfur exhibited an equal



FIGURE 1.1: Constituent Partitioning in MSW Incinerator Residues



FIGURE 1.2: Metal Partitioning in Bottom and Fly Ashes

distribution amongst the partitions, while zinc and lead were distributed equally in the fly and bottom ashes.

Similar partitioning relationships were observed during the NUS (1987) study as shown in Figure 1.2. Again, copper and iron were strongly favored in the bottom ash, while arsenic, cadmium, mercury, and zinc demonstrated their strongest affinity for the fly ash. To a slightly lesser extent, calcium, chromium, and nickel showed their strongest affiliation with the fly ash stream.

Dominant Mineral Phases

Understanding the mineralogy of MSW incinerator residues is important in predicting metal solubility and hence leachability. Numerous studies have been performed to characterize the prevailing solid phases in MSW incinerator residues. Methods used to study MSW incinerator residues include gravimetric/ titrimetric, x-ray diffraction (XRD), scanning electron microscopy (SEM), and dispersive analysis of x-ray (EDAX) techniques.

Several researchers report the predominant solid phase in fly and bottom ashes is silica oxide (Donnely & Jons, 1987; Griepink & Muntau, 1987; Hjelmar, 1987). Henry et al. (1987) noted similar findings, but also identified other solid phases existing in the fly ash matrix. Using XRD with Fourier Transform infrared techniques, they observed the

presence of various metal sulfates, chlorides, and carbonates.

In a more recent fly ash study, Ontiveros (1988) characterized the predominate crystalline surface species as complex sodium, calcium, and magnesium aluminum silicates in addition to silica oxides. He also identified a notable presence of sodium and potassium chlorides, calcium sulfates, and to a lesser extent titanium oxides.

<u>Metal Speciation</u>

Tessier et al (1979) designed a procedure for partitioning particulate metals into five general speciation categories. The five categories or fractions in the order of increasing metal availability were (1) exchangeable, (2) carbonate bound, (3) iron and manganese oxide bound, (4) organic matter bound, and (5) residual. Release of metals during Fraction (1) is commonly associated with changes in ionic composition of a leaching media, while Fraction (2) represents releases mainly due to pH changes. Metals released in fraction (3) and (4) leachates are commonly associated with shifts in oxidation-reduction conditions, and Fraction (5) represents those metals not expected to mobilize under typical environmental conditions due to their incorporation into the crystalline structure of the mineral. Tessier reported satisfactory accuracy by comparing the sum of sequentially extracted metals to their total

concentration measured in fluvial bottom sediments. Additionally, Tessier reported precision to be \pm 10% for replicate analyses (Tessier, 1979).

Several studies have used the sequential extraction procedures outlined by Tessier for characterization of MSW incinerator residues. Brindle et al (1987) classified cadmium, chromium, copper, nickel, lead, and zinc from a MSW fly ash. He found that lead and chromium were primarily associated with iron/ and manganese oxides (fraction 3), while all other metals released greater than 50% of their total content during Fractions (1) & (2) extractions.

Similarly, Eighmy et al (1989) performed a sequential chemical extraction on bottom ash samples generated from the same incinerator used in this research. Results indicated that cadmium, nickel, zinc, and lead were primarily affiliated with Fractions (3) and (5), while copper, chromium, and iron were predominately leached from the residual fraction.

Laboratory Leaching Studies

Numerous types of leaching tests can be used to estimate leachable contaminants from solid waste materials. Three types of tests in increasing complexity, cost, and realism are batch, column, and pilot leaching studies. These tests are often critical in assessing waste toxicity

and in providing necessary information for the formulation of waste management strategies. The choice of the appropriate leaching test is predicated on research objectives, availability of resources, and the timeliness of required results.

Batch Tests

Batch leaching tests are generally inexpensive to perform, simple to operate, and flexible in design and leaching protocols. The low costs of performing leach testing permits the increased replication of experiments, hence increasing the statistical significance of the results (Jackson et al., 1984). Similarly, the flexible nature of the tests permit the initial tailoring of leaching protocols and the opportunity to "fine tune" experiments based on data generated from previous batch analyses.

Standard batch leach tests were created for use in regulatory actions and waste classification objectives. Two such leach cests are the Extraction Procedure Toxicity Test (EP Toxicity) and Toxicity Characteristic Leaching Procedures (TCLP) test (EPA, 1982).

<u>EP Toxicity</u>. The EP Toxicity test is an agitated acetic acid extraction procedure conducted on waste forms subjected to a 20:1 liquid/ solid ratio and a pH environment of 5 \pm 0.2. Twenty-four hour leachate samples are filtered through a 0.45 um membrane and analyzed for National Primary

Drinking Water Standard (NPDWS) listed constituents. If the dissolved constituent concentrations measured in the 24 hour leachate samples exceed 100 times the regulatory limit for drinking water, the waste is assigned a hazardous rating (EPA, 1982). The maximum allowable concentrations according to EP Toxicity guidelines for present and proposed regulated constituents are listed in Table 2.1.

TCLP. The TCLP test is another regulatory tool originally devised to replace EP Toxicity. The tests are similar procedurally with the major difference being the method of agitation, strength of the leaching media, and duration. TCLP uses a rotary extraction vessel, a more aggressive acetic acid leaching media (pH = 2.88), and is conducted over a 18 hour period. Additionally, leachates extracted during the TCLP procedure are subjected to more extensive analyses than EP Toxicity leachates. Due to these mechanistic differences, the TCLP test appears to be more aggressive in extracting chromium, nickel, and arsenic, less aggressive for copper and zinc, and about the same for cadmium and lead (EPA, 1987).

Column Studies

Column studies have long been considered the best procedure for generating realistic leaching data in the laboratory. Column configurations and operating parameters vary with research objectives, but in general column studies

simulate the infiltration of rainwater through a unit volume of waste material to better estimate resulting leachate composition from landfilled residues.

No true standard column leaching test exists, which often makes direct comparisons of various column study results difficult. Miner et al. (1986) proposed that a standard method for column leaching be adopted using saturated up-flow conditions. In support of this, Miner claims 55% reproducibility using similarly configured column experiments. Other researchers argue that saturated conditions fail to model the true nature of landfill leaching, which predominately involves hysteric wetting and drying in response to precipitation events (Thompson, 1989; NUS, 1987). They proposed that column tests employing intermittent wetting more accurately model the unsaturated conditions which typify most landfill scenarios, and promotes a realistic downward dislocation of mobile metal fractions in a wave front.

Column and Batch Comparisons

Researchers have noted that batch and column leaching results yield essentially the same data (Hjelmar, 1987) Each test offers distinctive advantages in assessing leachability of constituents from waste forms. Batch tests have been shown to be quicker, more reproducible, simpler to operate, and less expensive than column studies (Jackson et

al., 1984). Column analyses are thought to be more representative of actual field leaching conditions, showing good correlations between field study and laboratory data in several MSW incinerator leaching studies (NUS, 1987; Hjelmar, 1989; Hjelmar, 1987). Hjelmar also notes that column studies are the only method that provides an assessment of the initial percolates generated from the leaching of wastes under low liquid/ solid ratios.

Reproducibility problems associated with column study results are typically related to channelling of the leaching media, non-uniform packing of residue, clogging, and biological growth within the columns (Cote & Constable, 1982).

Solidification/Stabilization Technology

Solidification/ stabilization (S/S) technology has been an integral part of hazardous waste management practices for years (Conner, 1979). Application of S/S principals to the handling of MSW incinerator residues is a more recent phenomenon. Solidification/ stabilization of a waste form involves mixing with one or more admixtures to produce a more ecologically acceptable product. Although solidification and stabilization are often referred to interchangeably, they are mutual exclusive processes. Solidification refers to those operations which improve the physical and handling characteristics of a waste through the

creation of a dimensionally stable monolith. Stabilization refers to processes by which a waste is converted to a more chemically stable form. Stabilization in essence, detoxifies a waste by limiting the mobility of its various constituents (Brown, 1984; Wiles, 1987).

Numerous S/S agents are available and used in the disposal of hazardous materials. Some examples of S/S agents which could be used in amending incinerator residues include: bentonite-cement (Clark, 1986; Cote & Hamilton, 1984), soluble silicate-cements (Cote & Hamilton, 1984), thermoplastic and organic polymers (EPA, 1979), fusion or glassification (OECD, 1977), or cement and lime based amendments (Bishop et al, 1985; Cote & Hamilton, 1984; Ormsby, 1989).

Many of these agents and amending procedures are proprietary in nature. This study focused on the solidification and stabilization of a MSW combined (bottom & fly) residues using the cement, lime, and thermoplastic (asphalt) agents, and therefore will restrict the review to previous studies involving these specific amendments.

Metal Immobilization Mechanisms

While many S/S studies have been conducted on a variety of wastes forms, few have focused solely on the S/S of MSW incinerator residues. Therefore, the discussion of immobilization mechanisms will be generic to all solidified/
stabilized wastes. Specific release mechanisms pertaining to metals commonly associated with MSW residues are discussed in detail (by metal) throughout Chapter 3.

Overview. In the 1970s, the focus of S/S practices shifted from the fabrication of dimensionally stable waste monoliths to the reduction of waste leachability. With the shift in focus came a shift in the research direction, spawning numerous studies in assessing leaching characteristics of various solidified/ stabilized materials. Research efforts continue aimed at defining the long term leaching behavior of the various solidified wastes in an effort to establish more prudent waste management strategies.

The overall leachability of a metal from a solidified/ stabilized waste form primarily depends on whether it remains dissolved in the pore system solution or is immobilized through a chemical reaction. Malone and Larson (1982) expanded on this simplistic interpretation by describing the chemical mechanisms which can effectively fix contaminants in a solidified/ stabilized waste form. They include the production of insoluble compounds, adsorption/ chemisorption, passivation of waste particles, and substitution in insoluble crystalline materials. The latter mechanism is important in the removal of constituents from natural geochemical systems, and may figure prominently in an on-going study involving the attenuation of metals from

MSW incinerator residue leachates by various soils (McHugh, unpublished). Passivation of the waste particle entails the formation of a precipitant on the particle's surfaces which isolates the particle from the bulk leaching media. These metal immobilization mechanisms occur only incidentally in cement and lime based processes (Cote, 1986). Since precipitation/ dissolution and adsorption/ desorption are the dominant metal immobilization mechanisms associated with solidified/ stabilized waste forms, they will be discussed in detail under the following sections.

<u>Cement & Lime Amended Wastes</u>. Cement and lime consist of insoluble non-toxic substances forming large skeletal structures. These structures are very porous and become impregnated with the waste upon mixing. The waste material and its metals are free to react with the various components of the cement and lime solidification/ stabilization agent. Due to the high pH associated with the cement and lime, the principal metal reactions involve the formation of insoluble metal hydroxide, carbonate and to a lesser extent sulfide precipitates (Cote, 1986).

Numerous researchers cite the high pH and buffering capacity of cement and lime as a significant barrier to metal mobility from MSW incinerator residues (Hjelmar, 1989; Cote, 1986; van der Sloot et al., 1987). The high pHs are also cited as a possible release mechanism for amphoteric

metals such as lead, nickel, and zinc (Cullinane & Jones, 1989).

In general, metal carbonates formed within the solidified/ stabilized matrix will be more stable than the metal hydroxides (Cote, 1986; Brown, 1984). Metal carbonate formation may be dominate in landfilled solidified/ stabilized waste forms due to the process of carbonation. Calleja (1980) describes carbonation as the neutralization and formation of metal carbonates resulting from the contact of the waste with groundwater under relatively high carbon dioxide partial pressures. The process of carbonation represents another major benefit of cement and lime waste stabilization.

Hjelmar (1989) estimates that long term leaching of landfilled MSW incinerator residues will eventually promote decreasing pH and increasing reducing potential. These conditions will favor the formation of metal sulfides which have solubilities several orders of magnitude lower than the metal hydroxides and carbonates (Lindsay, 1979). The probability that cement and lime amended wastes will experience the required erosion in the pH and oxidizing conditions necessary to promote metal sulfide precipitation is questionable given their high buffering capacities.

The formation of metal silicates is another potential metal immobilization mechanism associated with cement and lime S/S technology. Iler (1979) describes metal silicates

as non-stoichiometric compounds where the metal is coordinated to silanol groups (SiOH) in an amorphous polymerized silica matrix. The primary advantage of metal silicate formation is that the silica matrix remains relatively insoluble over a wide pH range (2 - 10).

Numerous researchers have recognized the importance of metal oxides surfaces as adsorption sites for various cationic species prevalent in MSW incinerator residues (Hjelmar, 1989; Eighmy, 1988; Kinneburgh et al., 1976). Cote (1986) explains that oxides of iron, aluminum, silicon, and manganese are strongly hydrolyzed in aqueous solutions and thus possess a net positive surface charge at low pH, and a net negative charge at higher pH. Adsorption of cations to the metal oxides surfaces takes place when the cations exhibit a higher pH than the metal oxide's zero point charge pH. Past studies have shown that this mechanism loses importance in more reduced environments (Theis & Richter, 1979; Feijel et al., 1988).

The most evident and often overlooked immobilization mechanism is the reduction of leachable surface area through the creation of a cement or lime amended waste monolith. Many leaching studies (including this one) undermine this beneficial aspect of S/S technology. Studies of cement monolith leaching indicate that the predominant metal release mechanism is the outward diffusion of constituents dissolved in the pore water systems (van der Sloot et al.,

1987; Cote, 1986). This typically occurs through contact with an acidic leaching media (e.g. groundwater). Metals solubilized at the monolith surface or leaching front, are subject to concentration gradients which force them both outward and inward. The inward and outward flux can serve to provide a leached layer along the surface which tends to protect the core of the solidified/ stabilized waste form direct contact with an ~cidic environment (Cote, 1986).

Asphalt Amended Wastes. As with most thermoplastic agent, asphalt binder immobilizes waste constituents through microencapsulation. Microencapsulation involves solidification of the individual waste particles as to prevents chemical interaction with the environment, and hence mobility of constituents. When adequate coverage of a waste is achieved, the leaching performance of asphalt based materials typically exceeds that of cement and lime based agents (Cullinane & Jones, 1989).

Future studies should enhance the current information base concerning metal mobility from solidified/ stabilized waste forms. A better mechanistic understanding of the factors affecting the release of contaminant will assist in the design of safer landfills and more realistic leaching tests.

Reuse Potential of S/S MSW Incinerator Residues

Reutilization of S/S MSW incinerator residues have

predominantly focused on road construction applications. Typically, the studies have been more concerned with the solidified/ stabilized waste product's structural properties and performance than with assessing the potential for environmental impact through constituent leaching. Past research efforts in the reuse of solidified/ stabilized waste forms in the construction industry are offered as examples of future potential.

<u>Cement Residue Material</u>. In a study conducted at the Valley Forge Laboratories, MSW incinerator residues were batched with varying proportions of portland type II cement (Collins & Ormsby, 1978). Several specimens, replacing 100% and 50% of the total aggregate requirement with MSW incinerator residues, were fabricated and tested. The authors cited several problems. In all batches, hydrogen gas generation was observed due to the reactions of cement alkalis with the free aluminum inherent in the residues. This resulted in a detrimental volume expansion and spalling in all test specimens (Collins & Ormsby, 1978).

Similar results were obtained in a feasibility study conducted at Notre Dame University involving the amendment of processed residues with portland cement concrete. Compressive strengths (28 day) were measured as high as 4000 psi in some residue amended concrete specimens. Despite the relatively high compressive strengths, the expansion and spalling exhibited by the specimens lead

researchers to conclude that the reutilization of cement amended residues may not be a very successful endeavor (Lauer & Leliaert, 1976).

Greater degree of success was realized for cement solidified residues in a study exploring its potential reuse in concrete masonry unit (CMU) or block fabrication. Various mix designs produced CMUs that satisfied ASTM C 90 criteria for grade N hollow load bearing units (Lauer & Leliaert, 1976).

Lime Residue Material. Lime amended MSW incinerator residues were tested as a base course material under the patented name of "Chempac". The amendment process involves the addition of 2 - 10% hydrated, quick, or flue lime to residue maintaining a maximum particle size of one inch and a minimum carbon content of 10%. These mixtures developed 28 day compressive strengths of over 300 psi when mixed at optimum moisture content (18 and 23%) and placed at 95% of the density determined according to ASTM D 1557 (Collins, 1978). Additionally, average California Bearing Ratio (CBR) values were determined to be 80, with high measures of approximately 120. Similar results were obtained from the testing of a lime-treated incinerator residues placed as a base course for a Chicago parking lot (Zimmerman & Gnaedinger, 1977).

<u>Asphalt Residue Material</u>. Incinerator residues were used as aggregate in the bituminous base course construction

of a test section road in Houston. The paving material was termed "Littercrete" and contained 89% aged residue, 2% hydrated lime (as an anti-stripping agent), and 9% AC-20 asphalt binder. All percentages were based on total specimen weight. The mix design was determined through optimization studies which tested specimens containing between 9 - 12% asphalt content. The Marshall stability for compacted laboratory samples of the 9% asphalt specimens averaged 1360 lbs at a flow of 19/100-inches, exceeding requirements for medium heavy traffic (Collins & Ormsby, 1979).

CHAPTER II

METHODS AND MATERIALS

Residue Amendment Procedure

<u>Residue Preparation</u>

The combined incinerator residue used in this study consisted of bottom and fly ashes generated through the combustion of municipal solid wastes (MSW) by the Lamprey Regional Solid Waste Cooperative Incinerator in Durham, New Hampshire. The mass burn facility (108 tons/day) is comprised of three Consumat^R modular starved air incineration units and two low pressure steam generating boilers. Incineration of MSW occurs in the lower starved air chamber at 1400°F to 1600°F. Hot gases from the combustion process are reignited in the upper chamber at 1800°F upon addition of excess air. Figure 2.1 illustrates typical MSW incineration within the two-stage Lamprey Facility.

The Lamprey Incinerator serves numerous surrounding municipalities which vary in population and industrial base. The major inputs into the facility are paper wastes and metal related materials. Typical characteristics of the raw waste stream are summarized in Table 2.1 (Beck, 1988).

The bottom and fly ashes were generated by the Lamprey facility over the same three months period (March - May,



FIGURE 2.1: MSW Incineration in a Two-Stage Mass Burn Facility

TABLE 2.1: Typical Composition of MSW at the Lamprey Regional Solid Waste Cooperative Incinerator

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Waste	Composition	
Paper		
Newsprint	9	
Corrugated	10	
Other	17	
Plastic	6	
Yard Waste	15	
Food Waste	15	
Metal		
Non-Ferrous	2	
Ferrous	6	
Glass	12	
Misc	8	

1988) prior to collection. These residues typify normal operation of the facility. The fly ash was collected from ductwork inside the facility, while the bottom ash was obtained from a temporary storage pile. Consequently, the bottom ash was exposed to a leaching environment, both in the incinerator quench tank and the storage pile, prior to its collection.

Both ashes were sieved to remove material greater than one-half inch, and mixed into a homogeneous blend of 92% bottom to 8% fly ash by volume. A sieve analysis of the resulting combined residue was performed with the results shown in Figure 2.2. The combined ash exhibited a well distributed profile. The effective size (d_{50}) was measured at 0.08 mm, while the uniformity coefficient (d_{60}/d_{10}) equaled 20.

Amendment Additions

The sieved combined residue was mixed with varying proportions of type II portland cement and hydrated lime in a commercially available 6 ft³ barrel mixer. The combined residue was also amended with varying proportions of AC-10 asphalt using standard Marshall Hot Mix Design Procedures (ASTM D 1559, 1988). Amended residue mix ratios and corresponding water/amendment ratios (W/a) are given in Table 2.2.



FIGURE 2.2: Combined Residue Grain Size Distribution

TABLE	2.2	: Amended	Residue	Mix	Rati	los
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Matrix	Mix Ratio	Percent _Addition_	<u>w/a Ratio</u>
Cement	1:8	11	2.41
	1:5	17	1.53
	1:2	33	0.74
Lime	1:19	5	6.0
	1:9	10	3.0
Asphalt	1:6.5	18	N/A
	1:4.5	22	N/A
	1:3.8	26	N/A

The selected mix ratios for the cement amended residues were based on typical cement additions in concrete mixes of a high, medium, and low strength. Similarly, the lime percentages were chosen to mimic the levels associated with incinerators using acid gas scrubbers, while the asphalt mix ratios were chosen based on workability and preliminary Marshall stability results.

Four-inch diameter by 8-inch high cylinders of each cement and lime sample matrix were fabricated and cured for 28 days at room temperature $(24 - 27^{\circ}C)$ and 100% relative humidity. The cylinders were tested for compressive strength at varying time intervals according to standard specifications (ASTM C 39, 1988). Compressive strength analyses were also performed on 2" x 2" x 2" cubes cut from various asphalt amended test specimens using a MK Diamond^R (Hawthorne, CA) Concrete Saw. Additionally, the Marshall parameters of stability and flow, as well as density-void analyses were performed on the asphalt test matrices and reported according to standard ASTM criteria (ASTM D 1559, 1988).

Column Leaching Study

The 17% cement and 5% lime matrices used in the column study were fabricated in $42"(w) \ge 64"(1) \ge 3.5"(d)$ forms, and cured as previously described. After curing, the cement

amended residue was crushed to less than one inch using a 6inch jaw crusher and sieved to remove particles greater than one-half inch. The 5% lime matrix was crushed by hand, and sieved. The resulting materials were analyzed for size gradation using procedures outlined in ASTM C 136 (1988). Figure 2.3 illustrates the particle size distribution of the crushed amended residues. The size distribution of the unamended residue (from Figure 2.2) is superimposed for comparative purposes.

The amending of the combined ash increased overall residue particle size and decreased the range of grain size distribution, as evidenced by the change in effective size and uniformity coefficients. The resulting amended material therefore would be expected to possess less desirable compaction properties than the unamended residue.

<u>Column Design</u>

Continuous leaching of the crushed amended and unamended control residues was accomplished in 60"(h) x 10"(dia) schedule 40 PVC columns run in a fully saturated, downflow configuration as shown in Figure 2.4. Sampling ports were drilled into the column at 12 inch intervals and also served as a means to displace entrapped air during saturation with the leaching media. Three and one-half inch washed sand and gravel filter packs were placed at the bottom of each column separated by a 1 mm mesh. The 17%



FIGURE 2.3: Amended Residue Grain Size Distribution



FIGURE 2.4: Typical Amended and Control Column Design

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cement amended residue, 5% lime amended residue, and residue control columns were prepared in duplicate. The control columns contained 26.7 kg dry weight of combined residue, while the cement and lime amended columns contained 36.1 kg and 38.4 kg of combined residue in their respective amended form.

The average packing density for the amended and control columns was 64 and 70 lbs/ft³, respectively. A pH 4.0 \pm 0.3 synthetic acid rain was used as the leaching media. The acid rain consisted of 0.5 mls of 1N H_2SO_4 and 0.125 mls of 0.157N HNO3 per liter of type II water. The leaching media was allowed to saturate the columns slowly until an eight inch head above the packed residues was achieved. Pumps controlled the column effluent flows at 0.13 ml/min, while the acid rain influents were maintained manually. The effluent flow rate corresponded to an infiltration rate of 53 inches per year. Continuous leaching of the cement and lime amended columns was conducted to final liquid/solid ratios (L/S) of 0.6 and 0.4, respectively, while the control columns achieved L/S of 0.9. The amended columns took 146 days to reach their respective L/S, while the unamended control columns ran for 159 days. All columns operated under the same climatic conditions, maintaining an average temperature of 25°C within the column laboratory.

Analytical Protocol

All analyses performed on the column leachate were conducted in accordance with Standard Methods (1985) and ASTM (1988) procedures. Analyses on discrete leachate samples taken throughout the duration of the study included: alkalinity, pH, oxidation-reduction potential (redox), and conductivity. Alkalinity was determined potentiometrically (to pH=4.5) using 1.6N H₂SO₄, 0.16N H₂SO₄, and 0.1N H₂SO₄ titrants for the cement amended, lime amended, and control leachates, respectively. Alkalinity and pH were determined using a Beckman-Altex^R meter with a Beckman^R Gel-Filled Combination Electrode. Similarly, an Orion MD SA250 Meter with an Orion Platinum Redox Electrode was used to determine the leachate's redox potential. Conductivity was measured using a YSI Temperature Compensating Meter.

Metal and anion concentrations were determined from composite samples and reported as discrete analyte concentrations for the given composite period's highest liquid/solid ratio (L/S). Metals analyses were conducted by the New Hampshire Materials Laboratory (Dover, NH) using inductively coupled argon plasma emission spectrometry (ICP) (USEPA Method 6010, 1986). Concentrations of calcium, aluminum, zinc, cadmium, copper, nickel, manganese, lead, and sodium were determined from composites samples filtered through a pre-rinsed (type II water) 0.22 um membrane and acidified using concentrated reagent grade HNO₃. All

glassware used in metal sample preparation was soaked for a minimum of four hours in 50% HNO₃, and rinsed 7 times with deionized water followed by two type II water rinses. Chloride, bromide, nitrate, phosphate, and sulfate concentrations were measured on a Dionex^R Model 4000i Ion Chromatograph (USEPA Method 300.0, 1984). The anion samples were also filtered through a pre-rinsed 0.22 um membrane and an additional cation exchange resin prior to injection to minimize fouling of the instrument's separator column.

Analyses for alkalinity, pH, redox, conductivity, and metal concentrations were also conducted on leachate samples withdrawn from the columns at varying heights via their sampling ports. The ports were fabricated from lengths of Tygon^R tubing and 60 ml Nalgene^R bottles. Small mouth bottles were used with openings just large enough to permit passage of the pH and redox electrodes. Using these bottles and analyzing leachate immediately after extraction from the column minimized the influence of atmospheric O_2 and CO_2 on leachate composition. The analyses were performed periodically in an attempt to understand possible chemical interactions evolving throughout the length of each amended and control column. All analytical procedures were accomplished on the discrete leachate samples by previously described methods.

Batch Leaching Studies

Each amended residue matrix was subjected to identical batch leaching experiments to compare their relative leachable metal fractions and kinetic release profiles under controlled pH conditions. The amended residues and unamended control residue were mixed and cured as previously described, but crushed in a Weber Bros & White (Hamilton, MI) Model S500 Pulverizer to produce a material passing a #50 sieve. Batch leaching experiments were only performed on the amended and unamended material that passed a #50 sieve (d=0.296 mm).

Experimental Design

Four liter Nalgene^R containers were filled with 190 g of pulverized/sieved residue and 3800 ml of pH 4 \pm 0.3 synthetic acid rain to produce a slurry with a L/S of 20:1. The slurry was mixed continuously at 410 RPMs to ensure constant contact between the residues and the leaching medium using a Heler Lab Mixer (Floral Park, NY) equipped with a 2"(1) x 1"(w) stainless steel paddle.

The pH levels within the batch reactor were controlled at 10, 8, and 6, by a Fisher Scientific (Medford, Ma) Computer Aided Titrator (CAT). The selected pH range was considered representative of the diverse ash landfill environments that residues may ultimately be exposed. The

CAT system maintained a static pH condition by delivering small, precise titrant quantities to the reactor based on potentiometric signals received from a pH probe submerged in the slurry. Titrant strength was optimized based on the residue's buffering capacity, the instrument's capabilities to control pH, and the experiment's design to maintain a constant L/S. Figure 2.5 represents a typical schematic of the batch leaching apparatus.

Leachate Sampling

Leachate samples were extracted from the reactor at predetermined time intervals to provide insights into the kinetics of metals release from the various amended and unamended residues at each respective pH level. Batch experiments conducted on the pulverized amended residues involved leachate withdrawal at 0, 30, 60, 120, and 240 minutes after achieving each desired pH condition. To provide a leaching study more representative of actual environmental leaching conditions, the residue and leaching medium were not changed between each pH level of the batch study. The CAT system was merely reconfigured to provide a static pH at the next lower level, and sampling continued over the same time intervals.

The decision to conduct the batch studies in this manner was based on the results of preliminary leaching experiments using the pulverized residue control material.



FIGURE 2.5: Batch Leaching Test Design

In these experiments, the effects of extended leaching (for 24 hours) and the replacement of residue material and leaching medium after each respective pH level was evaluated. In general, most metals released a significant portion of their leachable fraction within the first 240 minutes of exposure, making extended leaching unnecessary for the purposes of this research. Additionally, the trends and kinetic profiles appeared relatively unaffected by the continued reuse of residue material and leaching medium at each successive pH condition. The results of the preliminary leaching experiments involving the pulverized residue control material are presented in the Appendix.

Leachate sampling was accomplished through pre-drilled holes in the reactor cap using a plastic 60 ml syringe and 6-inch length of Tygon^R tubing. The 40 ml leachate sample was centrifuged at 10,000 RPM for 10 minutes to separate particulate matter from the supernatant. Supernatant was then withdrawn from the centrifuge tube and filtered through a pre-rinsed (type II water) 0.45 um membrane filter, acidified using reagent grade concentrated HNO_3 , and submitted for metals analysis to the commercial laboratory using ICP. Leachate sampling and preparation are summarized in Figure 2.6.



FIGURE 2.6: Preparation of Leachate Samples for ICP Analysis

OA/OC Protocol

The quality control/quality assurance plan (QA/QC) involved random replication of alkalinity, pH, redox, conductivity, metal and anion analyses to verify measured leachate trends and assess instrument precision. Additionally, numerous spiked leachate samples were submitted for analyses by Ion Chromatography (IC), and Inductively Coupled Plasma Spectrophotometry (ICP) as blind performance audits. The results of the blind audits were used to determine instrument detection limits and ascertain overall experimental accuracy. Incorporation of blind audits and random replication in the QA/QC Plan was imperative given the residue leachates' diverse chemical composition.

Instrument Detection Limits

The IC's detection capability was based on a correlation analysis of the area count results from numerous injections of reagent blanks and standards of known analyte concentrations. From the correlation equation, expected area counts (\hat{y}) were calculated and the difference between \hat{y} and the actual area counts were used to determine a standard deviation of the blank, or s_b . The detection limits were then calculated as two times s_b (Miller & Miller, 1984).

Detection capability for the ICP used in all metals analyses, were determined independently by the commercial laboratory. The detection limit for a specific metal was calculated as three times the average standard deviation of that metal's measured concentration in repeated reagent blank analyses.

The detection limits for the IC and ICP were based on the results obtained from the analysis of known standards. This ignores the possible effects of matrix interference, which may be significant given the diverse composition of the residue leachates. Table 2.3 outlines the detection limits used in this study for each analyte, as well as the accepted detection limits for ICP by the US Environmental Protection Agency (USEPA, 1982).

Analytical Performance Audits

Replicate measurements of alkalinity, pH, redox, conductivity, metal and anion concentrations were accomplished randomly on various leachate samples. The relative standard deviation (RSD) between replicates of each analyses fell within \pm 7.2%. According to analytical protocol, known concentration standards for each metal and anion were run after every sixth sample to evaluate instrument accuracy. The results of these accuracy checks and the IC/ICP performance audits are given in Figure 2.7 as average percent recovery.

	IC		ICP	
	Experimental		Experimental	USEPA
<u>Anion</u>	<u>DL (ug/l)</u>	Cation	_DL (ug/l)	<u>DL (ug/l)</u>
Cl	300	AL	52	45
Br	420	Cd	9	4
NO _z	170	Ca	100	10
PO,	520	Cu	20	6
so,	310	Fe	10	7
4		Pb	66	42
		Mn	2	2
		Ni	8	15
		Na	1000	29
		Zn	8	2

TABLE 2.3: IC & ICP Detection Limits

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FIGURE 2.7: Average Accuracy of Metal and Anion Analyses

The average precision and accuracy measured for metals analyses using ICP was acceptable according to EPA Method 6010 (1986). Similarly, the average percent recoveries and standard deviations on chloride and sulfate analyses was acceptable based on single operator accuracy and precision standards for surface water samples listed in EPA Method 300 (1984).

CHAPTER III

RESULTS AND DISCUSSION

<u>Material Properties of Unamended & Amended</u> <u>Combined MSW Incinerator Residues</u>

Characterizations of unamended and amended combined ash waste forms represented a significant portion of this research. The characterization studies provided insights into each amendment's potential to solidify and chemically bind with both toxic and benign constituents of combined MSW incinerator residue. Additionally, residue characterization studies were used to form the basis for initial predictions and interpretation of leachate composition generated during the numerous leaching experiments.

Combined Ash Characterization

The elemental composition of the sieved combined ash was determined through a total acid digestion (Tessier et al., 1979) of representative residue samples and subsequent analysis of the resulting leachate by ICP. Figure 3.1 represents the residue's iron, aluminum, zinc, sodium, lead, copper, manganese, nickel, and cadmium contents. For the metals analyzed, iron was the predominant lement in the combined residue with a concentration of over 40,000 ppm, while the least prevalent was cadmium at less than 70 ppm.



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FIGURE 3.1: Elemental Composition of the Combined Residue

The concentration levels of analyzed metals correspond with previous studies conducted on a similar combined MSW incinerator residue (EPA, 1987; Fletcher & Garrison, 1986). Aluminum, manganese, lead, and copper levels were similar to those cited in past analyses of the Lamprey facility's bottom ash stream, while zinc and cadmium levels in the combined residue were higher, reflecting the influence of fly ash on elemental composition (Eighmy et al., 1988). Lower nickel and iron contents were found in the combined residue than found in the Eighmy study and may reflect differences in digestion procedures, raw waste stream, or operation and efficiency of the Lamprey Incinerator.

The sieved combined residue was also characterized for moisture content, volatile organic content, specific gravity, and porosity as previously described. The results of the analyses are listed in Table 3.1.

The large pore structure of the combined ash readily adsorbed water molecules allowing moisture contents of 29% to exist on ash particles that appeared to be physically dry to the touch. Rigorous handling of the ash affected the pore structure and promoted the release of moisture which produced a slurry material without external water addition. Through numerous gravimetric tests, the saturated surface dry moisture content of the combined ash residue was determined to be 22%.

The volatile organic content of the combined residue

TABLE 3.1: Combined Residue Characterization

Moisture Content	27%
Volatile Organics (Std Methods 209 D)	17%
Specific Gravity (ASTM 128-73)	
Bulk, G _{sb} Saturated Surface	1.605
Dry Basis, G _{cont}	1.855
Effective, G	1.804
Apparent, G _{sa}	2.140
Porosity, C	0.250

was not determined directly, but based on gravimetric analyses of bottom ash samples taken from the quench tank prior to deposition at the disposal landfill. These determinations ignore possible organic content contributions from the fly ash stream, which have been reported to be significant in past studies (USEPA, 1987; Metzger, 1987). Nevertheless, the fly ash's overall contribution of organics to the combined residue was assumed to be negligible given its small relative fraction (i.e. 8%) in the combined residue's total composition.

Although the size gradation was different, the apparent specific gravity of the combined ash from the Lamprey facility was similar to that cited in previous studies involving MSW incinerator bottom ash (Haynes & Ledbetter, 1975; Hartlen & Elander, 1986). A better indicator of the true physical nature of the combined residue is evident through its bulk specific gravity of 1.605. Comparing this to the bulk specific gravity of 1.605. Comparing this to the bulk specific gravity of common rock or sand (2.60 or greater) illustrates the combined residue's light and fluffy character.

Behavior of Amended Residue Specimens

While batching the individual cement and lime amended matrices, round spherical pellets were formed ranging from 1/2" to 2" in diameter. The pellets were moist on the outside with dry centers. The incremental addition of water
caused the size of the pellets to increase, thus requiring manual reworking of the material to ensure an equal distribution of water and a homogeneous blend of amendment and combined ash. Similarly, the asphalt amended specimens required manual reworking and far greater asphalt addition than typical asphaltic concrete mixes to achieve solidification. The performance of the cement, lime, and asphalt amended matrices during batching probably stems from the combined ash's inherent variability, vast pore structure and the localized addition of the water or asphalt. In both cases, the sites along the ash particle's surface quickly adsorbed the applied water or asphalt before contact to internal pores or other ash particles could occur. The high surface-saturated dry moisture content and porosity measured for the combined ash were also indicators of the residue's large pore structures.

Compressive Strength Results

Eight different amended incinerator residue matrices were created and studied. The combined incinerator ash served as the only aggregate material in the amended residues and represented the common element between each of the separate amended waste forms. Various material strength tests performed on the amended residues provided qualitative and quantitative measures of each amendments' potential to encapsulate and solidify the combined ash. Additionally,

these measures provided insight into possible reuse application of the amended waste forms in the construction industry.

Several cement and lime amended residue cylindrical specimens were created for use in conventional compression analysis. During curing, the amended material expanded above the form's top edge approximately 1/8" to 3/8". Additionally, air released and small cracks appeared along the specimens' surface during the first several hours of curing. The amended specimen's behavior is consistent with that of air entrained concrete mixes where finely ground elemental aluminum powder is added to react with cement alkalies to produce hydrogen gas (Kosmatka & Panarese, 1988). The aluminum inherent in the combined residue appears to have reacted with the cement alkalies in much the same manner, thereby causing the expansion and surface cracking seen in the amended specimens. Reuse of similar amended residues in the construction industry may require special metal recovery practices for the removal of aluminum from the residue and/or compaction methods to minimize possible air entrained in the material's pore structure.

Figure 3.2 contains the results of the compressive strength tests conducted on the various cement, lime, and asphalt amended test specimens. As expected, increasing cement and lime content increased 28 day compressive strength. Of particular interest was the linear



FIGURE 3.2: Compressive Strength Results from Cement, Lime, and Asphalt Amended Test Specimens

relationship which developed between 28 day compressive strength and cement content. A one percent increase in cement amendment yielded an increased compressive strength of approximately 20 psi within the given cement addition range.

The figure illustrates a similar increasing strength relationship between lime amended specimens cured for 28 and 270 days. The 5% and 10% lime matrices increased compressive strength at an average rate of 0.34 and 0.06 psi/day, respectively. A slow, steady increase in strength over time is characteristic of a pozzolonic reaction.

The relatively low range of compressive strengths for the cement amended specimens was expected due to the lack of conventional course aggregate, the presence of organic matter in the matrix, and entrainment of air due to the aluminum/cement alkali reaction. These amended mixes were designed to meet MSW incinerator residue solidification/ stabilization requirements and were not intended to yield high strength materials. Past studies reported higher compressive strengths for concrete/ash specimens using incinerator residues as a replacement for a portion of the fine aggregate in a conventional concrete mix (Cook, 1983; van der Sloot et al., 1987). In this study, the only aggregate in both the cement and lime amended specimens was the combined residue, which made the mixes more like a mortar material than a concrete.

The compressive strengths of the lime amended specimens were also expected to be low due to the relatively weak pozzolonic activity of the residue, compounded with the small lime percentage in the amended matrix. According to Cullinane & Jones (1989), 20 - 30 % lime content is required to totally react with a true pozzolonic material. The chosen lime concentrations for each matrix were based on typical lime contents seen in the residues produced from incinerators using flue gas cleaning materials (Forrester, 1988).

For comparative purposes, the asphalt amended specimens were also tested for compressive strength, as shown in Figure 3.2. Conventional asphaltic concrete specimens containing asphalt additions ranging from 18% to 26% would be expected to produce a very pliable material, having little or no compressive strength. On the contrary, the asphalt amended specimens possessed significant compressive strength rivaling that of the cement specimens. Potential large scale use of a similar asphalt amended residue material would be economically prohibitive given the large volume of asphalt required to achieve solidification of the combined ash. Previous studies replacing a portion of the incinerator residue volume with conventional aggregate yielded a material of greater strength and stability requiring less asphalt addition and thus providing a more

economically feasible alternative (Haynes & Ledbetter, 1975; Patankar et al., 1979).

Marshall Mix Parameters

The asphalt amended specimens were subjected to typical Marshall mix analyses to compare standard conventional paving mixture performance with mixes incorporating MSW incinerator residue. The results of the Marshall mix analyses may offer insights into the feasibility of using asphalt as a solidification agent for MSW incinerator residues and serve as a reference for future mix design optimization studies. Principal Marshall mix design features for asphalt paving mixtures include bulk specific gravity determinations, density-void analyses, and stability-flow tests.

Specific Gravity. The specific gravity determinations for the AC-10 asphalt, unamended combined ash, and compacted asphalt amended test specimens serve as the basis of all Marshall mix calculations. Table 3.2 lists the average specific gravity of replicate test samples along with their associated standard deviations.

The specific gravity of the AC-10 asphalt used to solidify the combined ash was characteristic of commonly accepted asphalt cement. Significant differences existed between the residue's bulk, effective, and apparent specific gravities and the corresponding specific gravities typically

TABLE 3.2: Average Specific Gravity Determinations For Asphalt Amended Test Specimens

	AC-10 Asphalt	Combined Ash	18% Asphalt Comp Uncomp	22% Asphalt Comp Uncomp	26% Asphalt Comp Uncomp
Specific Gravity, G _b (ASTM D 70)	1.0222				
Specific Gravity (ASTM 128-73)					
Bulk, G		1.605	1.38	1.32	1.42
+ SD * SD		0.007	0.141	0.014	••
Effective, G		1.804	1.76	1.69	1.97
+ SD Se		0.149	0.184	0.035	••
Apparent, G.		2.14	1.68	1.56	1.71
± SD		0.014	0.134	0.028	••
Specific Gravity					
(ASTH 2726)					
Bulk, G.,			1.311	1.373	1.471
+ SD mD			0.024	0.018	0.042
Maximum, G			1.556	1.473	1.587
± SD			0.120	0.021	

Comp=Compacted Specimen Uncomp=Uncompacted Specimen

associated with coarse and fine aggregates (Asphalt Institute, 1974). These differences suggest an increase in the number and size of voids in the MSW incinerator residue material, compared with that of conventional aggregate. The combined residue's vast pore structure not only affected specific gravity, but the other Marshall mix parameters included in the density-void and stability-flow analyses.

The specific gravities listed for the asphalt amended matrices were determined on compacted specimens that were not coated with paraffin prior to analysis. Omission of this step from the procedure may have affected the data's accuracy due to the specimens' porous nature and the inability to seal the residue with even 26% asphalt addition. Therefore, the results are included as relative measures from which amended asphalt matrices may be compared with each other since each specimen was treated identically during the analyses.

Density-Void Analyses. The primary properties determined in any density-void analysis are the compacted specimen's density, voids in the mineral aggregate (VMA), and air voids. The density of a compacted asphalt specimen, reported as pounds per cubic foot (pcf), is determined by multiplying its bulk specific gravity by the density of water. VMA represents the volume within the compacted specimen which is not occupied by aggregate. It is typically expressed as a percent of the total specimen's

volume. Air voids are also expressed as a percent of the total volume of the compacted specimen and represents that volume not occupied by asphalt or aggregate. In Figure 3.3, the density-void properties of the 18%, 22% and 26% compacted asphalt amended specimens are superimposed over density-void properties determined for conventional asphalt paving mixtures of medium strength (Asphalt Institute, 1974). The comparison of these trends highlight some major obstacles to the reutilization of similarly composed asphalt amended residues in the road construction industry.

All the amended residues' density-void properties deviated considerably from the ranges shown for typical conventional asphalt mixes. The asphalt content required to solidify the MSW incinerator residue was over four times that required in conventional materials, while the corresponding unit weights were approximately half those typically associated with standard mix designs. Similarly, the VMA and air voids measured in the asphalt specimens were considerably higher for the residue mixes than for mixes containing standard aggregate blends.

The trends demonstrated by residue and conventional paving materials also deviated considerably. A comparison of the unit weight plots showed that additional asphalt is required before maximum unit weight can be determined for the MSW incinerator residue mixes. The plot of percent air voids in the asphalt amended residues followed the expected





FIGURE 3.3: Density-Void Properties of Compacted Asphalt Amended Specimens

trend at first, then unexpectedly showed an increase in air voids with a corresponding increase in asphalt. Similarly, the sharp decrease seen in the VMA plot for the residue materials also suggested an increase in air voids with increasing addition of asphalt. These inconsistent responses to the addition of asphalt indicate high variability in the volume of asphalt absorbed by the residue due to the porous and variable nature of the combined ash.

Stability-Flow Tests. Stability of an asphalt paving mix reflects its ability to resist deformation from imposed loads. Stability is a relative measure of an asphalt matrix's strength resulting primarily from the internal friction among the aggregate and cohesion provided by the asphalt binder (Asphalt Institute, 1974). The stability of a test specimen is its maximum load resistance developed at 140°F. Flow is also a relative measure of strength, and is taken as the difference between the total movement or deformation of a test specimen at no load and maximum load. Flow is reported in units of 0.01 inches, and is measured simultaneously during stability analysis. The results of the stability-flow tests for the various asphalt amended matrices, along with typical stability-flow plots associated with conventional mixes, are shown in Figure 3.4.

The stability range determined for the asphalt amended residues were comparable to conventional mixes containing considerably less asphalt binder. A maximum Marshall





FIGURE 3.4: Stability-Flow Test Results from Compacted Asphalt Amended Specimens

stability of approximately 2000 lbs was achieved for MSW incinerator residue mixes containing 23% asphalt content. The flow ranges and trends measured during stability testing of the residue mixes differed from flows reported for conventional mixes. Typically, conventional paving mix flows increase almost exponential with small increases in asphalt content (Asphalt Institute, 1974). The flow trend determined for the residue mixes showed little similarity, appearing unaffected by significant increases of asphalt binder.

Data Interpretation. General observations concerning density-void and stability-flow results are possible, assuming the error induced by the improper preparation of the residue test specimens had a relative effect. The difference in unit weight, VMA, air voids, and flow trends between the residue and conventional mixes may all be due to the influence of the residue's porous nature, and its propensity to readily absorb asphalt binder. The irregular manner in which asphalt was absorbed by the residue particles induced variability in the tests results and inconsistencies in the VMA and air void trends. Additionally, the residue mixes' failure to achieve maximum density over the wide range of asphalt contents tested, and the inability to affect flow through asphalt addition all suggests that many of the residues pores were left uncovered by binder material. The inability to cover and seal the

residue particles may have far reaching consequences if the amended residues were subjected to leaching.

Although a 2000 lbs Marshall stability measured at a flow of 23 may be acceptable for most conventional paving applications, its achievement comes at a considerable cost. Improvements in mix designs to include conventional aggregate material must be pursued if serious consideration is to be given to large scale utilization of MSW incinerator residues in asphalt paving applications.

Ideally, the optimum asphalt content for a mix can be determined from the density-void and stability-flow plots. It is determined to be that asphalt content yielding the highest stability, maximum density, and medium air void The failure to paraffin coat the compacted asphalt content. amended specimens obviously affected the validity of the data, therefore a recommendation of the optimum asphalt content for use with MSW incinerator residues can not be provided. Due to the porosity inherent in the combined residue, future studies should include the coating of asphalt amended test specimens with paraffin prior to contact with water in the specific gravity analyses. In considering possible reuse applications in construction for asphalt amended incinerator residues, future tests should explore the enrichment of the matrix with conventional aggregate. This should limit the quantity of asphalt binder

required, increase stability, and permit better estimation of optimum mix design for amended MSW incinerator residues.

Column Leachate Analyses

Column leaching studies were conducted on duplicate columns of 17% cement amended, and 5% lime amended, and unamended MSW incinerator residues (control). The amended residues were crushed and sieved prior to column loading to simulate a worse-case leaching scenario. Leaching kinetics of monolith-like waste forms would likely result in much lower constituent concentrations in the leachate. Further, no adjustments were made to any leachate parameters due to the slight increased mass of combined residue present in the amended columns. The difference provides a more conservative estimate of metal leachability from the amended residues relative to the control residues.

Environmental Parameters

The environmental conditions surrounding any waste material have a profound effect on leachability. Several researchers have attributed metal leachability to the waste forms' physical and chemical properties as well as the characteristics of the hydraulic regime surrounding the waste (Bridle et al. 1987; Cote, 1986). Some of the environmental characteristics of importance in predicting leaching potential of a waste form are alkalinity, pH, redox, and conductivity (Constable et al, 1984). These parameters were measured throughout the column study and are reported in Figures 3.5 - 3.8 as a function of liquid/solid ratio (L/S).

Alkalinity & pH. Previous studies indicate leaching of heavy metals from a solidified/stabilized waste form is governed by the amount of alkalinity present in that waste (Bishop et al, 1985). The higher the buffering capacity provided by the amending agent, the more resistant the stabilized waste form will be to attack from acidic rain or soil water. However, extremely buffered systems containing caustic alkalinity can cause the release of metals that are amphoteric and soluble at high pH levels. Figure 3.5 shows the trends in leachate alkalinity from the 100% combined residue (control), 17% cement, and 5% lime columns.

The alkalinity of the combined residue and lime amended leachates increased slightly at first, then remained relatively constant throughout the study. The cement amended leachates demonstrated a sharp increase in alkalinity through a L/S of 0.22, followed by a continual, more gradual increase through the rest of the leaching period. The pH (Figure 3.6) of the respective leachates also followed similar trends. This suggests buffering of the acidic leaching media occurs rapidly to pH and alkalinity levels characteristic of the waste form itself.



FIGURE 3.5: Alkalinity Levels Measured in Amended and Control Column Effluent Leachates



FIGURE 3.6: pH Levels Measured in Amended and Control Column Effluent Leachates

An apparent anomaly exists between the alkalinities measured in the residue control and lime amended leachates. The addition of 5% lime to the residue caused a decrease in alkalinity and pH. Attention was focused on the equilibrium conditions with respect to $CaCO_{3(s)}$ in the lime and control columns in an effort to explain this phenomenon. The saturation or Langelier Index (LI) is a common indicator of $CaCO_{3(s)}$ equilibria of a system, and is defined as the difference between the actual (measured) pH of an aqueous solution, and the theoretical pH in equilibrium with CaCO_{3(s)} (Snoeyink & Jenkins, 1980). Analysis using the average pH, temperature, ionic strength, calcium, and bicarbonate concentrations found in the control and lime amended column leachates revealed a slightly oversaturated condition existing in the control column with respect to $CaCO_{3(s)}$, while the lime column leachate was strongly undersaturated. Oversaturated systems tend to precipitate $CaCO_{3(s)}$, therefore the reaction shown in Equation 1 is thought to predominate in the control column leachate.

$$Ca^{2^{+}} + HCO_{3^{-}} < --> CaCO_{3(a)} + H^{+}$$
 [Eq 1]

The undersaturated conditions in the lime amended column suggests that $CaCO_{3(s)}$ is dissolved via the same reaction in the reverse direction. This may not be entirely true since the addition of lime in the form of Ca(OH)₂ significantly

increases the stoichiometric quantities of calcium and hydroxide ions, therefore encouraging further precipitation of $CaCO_{3(a)}$ according to Equation 2.

$$2Ca^{2^{+}} + OH^{-} + 2HCO_{3}^{-->} 2CaCO_{3(s)} + H_{2}O + H^{+}$$
 [Eq 2]

The likelihood of Equation 2 proceeding as written in the lime amended columns was assessed by calculating its change in Gibbs free energy (ΔG_{RXW}) at non-standard conditions. Average values of temperature (25°C), ionic strength (0.32), pH (6.92), calcium (422 mg/l) and bicarbonate (30.90 mg CaCO₃/l) concentrations measured in the lime columns throughout the study were used in the calculation. The bicarbonate concentration was taken as the average total alkalinity (to pH 4.5 ± 0.1) measured in the lime amended leachates. The assumption that all the alkalinity was in the form of bicarbonate was appropriate due to the leachates' average measured pH of 6.92 (Snoeyink & Jenkins, 1980).

The change in Gibbs free energy for Equation 2 was $\Delta G_{RXN} = -7.17$ kcal/mole, suggesting that the reaction has a strong tendency to proceed in the forward direction spontaneously. This was expected given the large negative LI (LI = -1.24) associated with the lime amended leachate. Since Ca(OH)₂ is more soluble than CaCO_{3(s)}, the lime columns' corrosive environment may have preferentially promoted the dissolution of $Ca(OH)_2$, which in turn initiated additional precipitation of $CaCO_3$. The theoretical reaction therefore explains the decrease in alkalinity due to the consumption of bicarbonate ions and also the decrease in pH due to the release of excess protons.

More extensive study should be performed to better ascertain the validity of this hypothesis. The LI and Gibbs free energy analyses are simple approaches used to explain a very complex system. Undoubtedly, the precipitation/ dissolution of other mineral phases may play a significant role in the overall chemistry prevailing in both the control and lime columns. Future studies should examine the chamended and lime amended residues using X-ray diffraction. If the predominating reactions in the control and lime amended columns are as presented in this report, CaCO₃ (calcite) should be present in much higher concentrations in the lime amended residue than in the unamended residue.

The pH ranges for the control leachates were similar to those cited in the literature for incinerator bottom ash (Eighmy et al, 1988). This was expected given the composition of the combined ash (i.e. 92% bottom to 8% fly ash). The control leachate alkalinities exceeded published data by a factor of 10 (Hjelmar, 1987; Cundari & Lauria, 1987; Eighmy et al, 1988). The lack of agreement may be due to variations in the raw waste streams or operating procedures at the various incinerator facilities cited.

Oxidation-Reduction Potential. In general, all redox measurements experienced a degree of variability not only with time, but also between replicate columns. Redox fluctuations are not uncharacteristic of platinum probe redox measurements, especially in solutions containing such wide ranging constituents such as MSW incinerator residue leachates (Eighmy et al, 1988). Lindberg and Runnells (1984) cite that of the thousands of E_h measurements reported for natural waters, very few are valid on a quantitative, Nerstian basis, due to the inherent disequilibrium in systems containing multiple redox couples. Figure 3.7 illustrates the redox potential measured in each of the leachates.

Variability notwithstanding, distinctive redox trends between each of the leachates are apparent. The cement amended leachates were consistently more reduced than the lime amended and control leachates. Possible redox couples occurring in the cement leachate include $SO_4^{2^-}/H_2S$, CO_2/CH_4 , and Fe^{3^+}/Fe^{2^+} (Drever, 1988). Additionally, the production of hydrogen gas via the previously described curing reaction involving cement alkalis and residue associated aluminum may have also contributed to the cement leachate's lower E_h through the release of electrons as depicted in Equation 3 (Kosmatka & Panarese, 1988).

 $\frac{1}{2}H_{2(q)} < --> H^{+} + e^{-}$ Eq 3



FIGURE 3.7: Redox Levels Measured in Amended and Control Column Effluent Leachates

A reduced environment, coupled with the high pH and alkalinity exhibited by the cement amended leachate should render most metals immobile and resistant to environmental leaching from the matrix (Cote et al, 1986).

The average redox measured in the control leachates corresponds to redox levels seen in a previous study of the Lamprey Incinerator bottom ash (Eighmy et al., 1988). The measured E_h of both the control and lime leachate systems suggest the major redox reactions involve Fe^{3+}/Fe^{2+} and MnO_2/Mn^{2+} (Drever, 1988).

More reliable redox data, in concert with pH and alkalinity, should enable better prediction of constituent leachability from specific waste forms and also lend insights into the metal speciations. Given the difficulties associated with E_h determinations in these complex leachate systems, future studies should consider direct analyses of redox sensitive species like oxygen, sulfides, and/or methane to better indicate the prevailing redox conditions.

<u>Conductivity</u>. Conductivity is a measure of an aqueous solution's total dissolved ion concentration. Figure 3.8 illustrates the measured leachate conductivities for the cement, lime, and control columns. The cement amended leachate demonstrated a relatively low and consistent conductivity compared to the lime amended and control leachates. This may be due to adsorption of dissolved solids to the cement paste matrix and the propensity of the



FIGURE 3.8: Conductivity Levels Measured in Amended and Control Column Effluent Leachates

silicates in the cement to form insoluble complex precipitates that deposit throughout the column (Cook, 1988). As seen by the wide error bars in Figure 3.8, the replicate lime columns demonstrated significantly different leachate conductivities. Initially, channelling in lime column #2 was suspected as the cause of this variability. Although this may have been a factor, the principal source of the variability was probably due to a non-homogeneous mixture of the combined residue with respect to sulfate concentrations (see Figure 3.12).

Leachate Anion Concentrations

Anion levels within the leachates were determined to assist in identifying possible metal speciations, and to provide general indications of potential microbial activity over time. The anions of interest within the composite column leachate samples were chlorides, sulfates, nitrates, phosphates, and bromides.

<u>Chloride & Sulfate Levels</u>. Chloride and sulfate concentrations differed dramatically between the unamended control and amended leachates, as shown in Figures 3.9 & 3.10. The 17% cement and 5% lime addition to the combined residue appears to have immobilized a significant fraction of soluble chloride and sulfate. More important is the implication that the amendments may be effective in



FIGURE 3.9: Average Chloride Concentrations Measured in Amended and Control Column Effluent Leachates



FIGURE 3.10: Average Sulfate Concentrations Measured in Amended and Control Column Effluent Leachates

solidifying and/or stabilizing potentially toxic metals speciated as chloride and sulfate salts.

Wide ranging variability between replicate lime and control leachates existed with respect to sulfate, and to a lesser extent, chloride concentrations. Replicate cement amended leachates showed chloride variation in the initial stages of the study, while sulfate concentrations were much more repeatable. Both anions exhibited conservative natures by demonstrating decreasing concentrations with increasing liquid/solid ratio in all the leachates tested. The relative concentrations and parallel trends of chloride, sulfate, and sodium (Figure 3.17) in the amended and unamended leachates suggests that much of their leachable fractions may be in the form of sodium salts.

Nitrate, Phosphate, and Bromide Levels. Each leachate sample was diluted to ensure chloride and sulfate concentrations remained within the linear range of the analytical instrument. Unfortunately, the detection limits for the other anions of interest increased accordingly, and their presence in the leachates became increasingly difficult to determine. Numerous attempts to determine nitrate, phosphate, and bromide concentrations on less dilute leachate samples were also unsuccessful due to chloride and sulfate interference. The limits of detections and detectable concentrations for nitrate, phosphate, and

bromide for the amended and unamended leachates are listed in Table 3.3.

Nitrate levels in all leachates were highly variable. Much of the variability stems from the use of nitric acid to prepare glassware used in the analyses. Extensive type II water rinsing of all acid washed glassware limited the degree of nitrate variability, but in itself, was unable to completely arrest the problem.

Soluble phosphate concentrations in all the leachates fell below detectable levels in nearly every analysis. Similarly, bromide levels in both the cement and lime amended leachates were undetectable, but significant bromide concentrations were seen in the unamended control leachate. The variability associated with the detection of bromide was far less than that of the nitrate analysis, with the control leachate exhibiting detectable bromide concentrations ranging from approximately 990 to 1200 ppm. These results suggest that the 17% cement and 5% lime amendments were successful in immobilizing bromide associated with the combined MSW incinerator residues.

Metal Leachate Levels

The metals selected for analysis in this research were based primarily on the results of studies involving MSW incinerator residues similar to those generated at the Lamprey facility, and on existing/proposed drinking water

Cement Amended Leachates										
L/S Ratio	NO3	SD	P04=	SD	8r [°]	SD				
0.009	6.2		< 104		< 84					
0.015	< 34		< 104		< 84					
0.031	< 68		< 208		< 168	••				
0.066	722.2	925.18	< 208		< 168					
0.108	343.8	389.97	< 208		< 168					
0.189	< 68		< 208	••	< 168	••				
0.260	< 49		148.6		< 120					
0.335	< 49		< 148		< 120					
0.388	< 43		< 130		< 105	••				
0.497	130.7		< 148		< 120	••				
0.571	< 49		< 148	* -	< 120					
0.623	< 43	••	< 130		< 105					
	Li	me Amended	Leachates							
L/S Ratio	NOz	SD	PO ₄ =	SD	Br	SD				
<u> </u>										
0 011	123.4	65 97	< 520		< 420					
0.017	< 68		< 208	••	< 168					
0.034	< 170		< 520		< 420					
0.067	595.0	453.6	< 520	•-	< 420	•-				
0.110	716.0	772.2	< 520	••	< 420					
0, 192	442.3	235.8	< 520		< 420					
0.267	< 85		< 260		< 210					
0.342	< 85		< 260	••	< 210	••				
0.395	< 85		< 260		< 210					
	11									
L/S Ratio	NO-	sD	PO, ⁼	SD	8r ⁻	SD				
0.015	< 636		< 104		1196	333 7				
0.015	2726		< 104		1108					
0.021	908	384 67	< 104		< 140					
0.029	974	225 65	< 104		1070	24.04				
0.098	4452		< 104	••	1116					
D. 11D	1345	1002.7	< 104		1070					
0.124	1452	1154.0	< 104		1096					
0 153	3140	•-	< 104	••	1018					
0.155	1744	••	< 104	••	< 140	••				
0.100	6110	••	< 104	••	< 140	••				
0.174	3950		< 104		< 140	••				
0.223	۵۲۶۵ ۵۶۸ >		< 104		007	62 21				
0.000	× 030		~ 104		776	JE . 2.				

TABLE 3.3: Nitrate, Phosphate, and Bromide Detectable Limits and Concentrations (ppm)

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standards established by the US Environmental Protection Agency (USEPA). More specifically, lead, nickel, zinc, aluminum, cadmium, and copper were chosen due to their potentially deleterious impact on both plant and animal life if released to the environment in significant concentrations. Calcium was selected for metals analysis because of its high relative concentration in the cement and lime amendments and the combined residue, while iron, manganese, and sodium were chosen to provide insights into the chemistry of the unamended and amended leaching systems.

Lead Concentrations. Lead concentrations in the residue control leachates were measured at trace levels throughout the duration of the column study. This is consistent with recent leaching studies which cite maximum lead concentrations for combined incinerator residue leachates at approximately 0.1 ppm (Eighmy et al, 1988; Hjelmar, 1987). The low lead concentrations found in the leachates confirms lead relative immobility under naturally occurring pH and redox conditions.

A dramatic increase in lead for the cement amended leachate (9-15 ppm) occurred at a L/S of 0.25 as shown in Figure 3.11. At this L/S, the average pH, alkalinity, redox, and ionic strength were measured to be 11.6, 1835 mg $CaCO_3/1$, -40 mv, and 0.2, respectively. Lead releases under these environmental conditions could have been associated with the reduction of lead chromates adsorbed to the



FIGURE 3.11: Average Lead Concentrations Measured in Amended and Control Column Effluent Leachates

surfaces of iron oxides, as well as the dissolution of various lead mineral phases (e.g. PbSO₄, Pb(OH), & PbCO₃) at high pH levels (Theis & Richter, 1979; Kinniburgh et al, 1976). Hem (1977) concluded that lead carbonates and basic lead carbonates are potential controls for lead solubility, especially in alkaline environments. While some dissolution of PbCO₃ and Pb(OH), may have occurred, the prevailing sulfate concentrations and ionic strength favors the dissolution of PbSO, as the primary lead release mechanism. This was evidenced by the relative agreement between measured lead concentrations (12 ppm) and predicted levels (3 ppm) associated with lead sulfate dissolution. The amphoteric nature of lead suggests that cement stabilized residues may leach lead in environmentally significant concentrations especially when the integrity of the monolith has been compromised.

Nickel & Zinc Concentrations. Trace concentrations of nickel (Figure 3.12) and zinc (Figure 3.13) were measured in all column samples analyzed during the study. The nickel and zinc concentrations determined for the combined residue leachates were consistent with levels cited in the literature (Cundari & Lauria, 1987; Hjelmar, 1987). Initial nickel and zinc levels in each of the column leachates were highly variable. Variability appeared to decrease significantly in the unamended control and cement amended leachates with increasing liquid/solid ratio.



FIGURE 3.12: Average Nickel Concentrations Measured in Amended and Control Column Effluent Leachates



FIGURE 3.13: Average Zinc Concentrations Measured in Amended and Control Column Effluent Leachates

The levels of zinc measured throughout the column study did not appear to differ significantly between the unamended and amended leachates. Conversely, nickel concentrations in the cement amended leachate samples exceeded the control and lime amended leachates by approximately 0.7 ppm at liquid/solid ratios greater than 0.2. Given the cement leachate's environmental conditions, the increased nickel release was probably due to the dissolution of nickel carbonates and nickel hydroxides. Additional nickel release may be attributed to its adsorption to iron oxide surfaces and subsequent release under reducing conditions (Theis & Richter, 1979; Feijel et al. 1988).

Aluminum Concentrations. Aluminum concentrations in the cement amended leachates varied significantly from those found in the control and lime amended leachates. The release of aluminum from the amended and unamended leachates is illustrated in Figure 3.14. The relative insolubility of aluminum in the control and lime amended columns was expected since their pH conditions (7.7 & 6.9) were near the point of minimum hydroxide solubility (EPRI, 1984). Conversely, the cement amended columns operated within a pH range that favored the formation and subsequent dissolution of negative hydroxy-complexes of aluminum (Stumm & Morgan, 1981). The dissolution of aluminum at pH of nine or greater, was confirmed through separate batch leaching experiments conducted on no fewer than nine different



FIGURE 3.14: Average Aluminum Concentrations Measured in Amended and Control Column Effluent Leachates

combined residue matrices. In the batch study, all unamended and amended residues were subjected to pH environments of 10, 8, and 6. The leaching of aluminum occurred only when a controlled pH 10 condition was applied. No soluble aluminum was detected at the pH conditions of 8 and 6 for any of the residues tested (see Figure 3.37). If the release of aluminum is solely a function of pH, solidification/ stabilization using only type II portland cement may present problems given the high alkalinity and pH inherent in the resulting waste form. Future research exploring methods for immobilizing aluminum from cement amended residues is warranted especially considering the recent concern about possible links between aluminum and various neurological disorders (Waldbott, 1973; Miller et al, 1984).

Calcium Concentrations. Significant calcium (Figure 3.15) concentrations were measured in the control and lime amended leachates, while the cement amended leachate exhibited trace levels of calcium for the majority of the study. The release of calcium from the three waste forms was the mirror image of the release patterns seen for aluminum. Complex adsorption/desorption reactions may explain these release trends. Previous studies have shown that aluminum is more strongly bound than calcium to cation exchange sites of layer lattice silicates (McBride & Bloom, 1977; Kozak & Huang, 1971). As a test, equivalent


FIGURE 3.15: Average Calcium Concentrations Measured in Amended and Control Column Effluent Leachates

concentrations were determined using average calcium and aluminum levels measured in the control and cement leachates. The calculation yielded calcium and aluminum equivalent concentrations of 20 ueq/l, and 22 ueq/l, respectively. The relative agreement of these values strongly suggests that calcium and aluminum participate in an ion exchange process in each of the leachates tested. The mobility of aluminum in the cement amended leachate does not necessarily negate this hypotheses. The dominance of aluminum precipitation/dissolution chemistry at the high pH levels measured in the cement column or the selective exchange between potassium may have been the controlling aluminum release mechanism (Pleysier et al., 1979; Coulter 1969).

All column leachates exhibited classic behavior with respect to calcium carbonate solubility. Samples taken during initial leaching in the cement columns maintained average calcium concentrations of 30 ppm, while samples drawn after L/S of 0.1 measured only trace levels of calcium. The pH conditions initially and at liquid/solid ratio of 0.1, was 9.3 and 10.1, respectively. Similarly, calcium concentrations in the control and lime amended leachates differed only slightly through most of the study, tracing the established pH trend previously illustrated in Figure 3.6.

Manganese Concentrations. Manganese was selected as a research parameter due to its highly reactive nature and strong dependence on the environmental parameters of redox and pH. This dependence helped in forming hypotheses concerning its dissolution/ precipitation tendencies, as well as the chemistry of other metals commonly associated with MSW incinerator residues.

More than any previous metal, manganese concentrations varied distinctly between the unamended control leachate and the amended leachates, as well as between each of the amended leachates. Figure 3.16 demonstrates the relative release trends of manganese in the unamended and amended combined residues. Significant attenuation of manganese was accomplished by the cement amendment, while the lime amendment demonstrated manganese immobilization for L/S greater than 0.18. The average redox and pH conditions measured in the cement amended leachates were -40 mv, and 11.6, while the control and lime leachates possessed average redox values of 300 and 100 mv, and average pHs of 7.79, and 6.92. Under the environmental conditions described, mineral phases of manganese (e.g. MnO₂) should prevail in the cement amended leachates, while manganese release in the control and lime amended leachates favors dissolution of manganese in its elemental form (Stumm & Morgan, 1981). Therefore, the differences seen in manganese levels appear to be directly attributable to the pH and redox conditions



FIGURE 3.16: Average Manganese Concentrations Measured in Amended and Control Column Effluent Leachates

prevailing in each of the respective leachates. This was expected based on the established behavior of manganese in the published literature.

Sodium Concentrations. Sodium is considered a conservative cation due to its nonreactive nature (Lindsay, 1979). The assessment of sodium levels in the various leachates served as a relative measure of each amendment's potential to effectively solidify MSW incinerator residues.

Similar release trends were seen with respect to sodium as were previously demonstrated with manganese. Figure 3.17 illustrates the average sodium concentrations associated with each column leachate. Sodium demonstrated a gradually decreasing release with increasing liquid/solid ratio in all leachates analyzed. This "washing out" trend was expected given the conservative nature of sodium. The cement and lime amended leachates experienced significantly lower average sodium concentrations than were measured in the residue control leachates. No significant difference between sodium concentration in the cement and lime amended leachates was apparent due to the high variability associated with replicate lime columns.

Batch leaching experiments performed on the unamended and amended residues also showed a significant difference between sodium concentrations in the control leachate and in the amended leachates (see Figure 3.43). More than half of the total available sodium was successfully attenuated



FIGURE 3.17: Average Sodium Concentrations Measured in Amended and Control Column Effluent Leachates

through encapsulation of sodium in the cement and lime paste in both the column and batch leaching tests. The results clearly indicate that both the cement and lime amendments effectively solidify MSW incinerator residues and inhibit the release of large leachable fractions of sodium to the environment. The ability to immobilize even small percentages of the total leachable sodium fraction from incinerator related residues may prove significant given the proposed action by the EPA to regulate sodium concentrations in drinking water.

<u>Cadmium, Copper, and Iron Concentrations</u>. Other metals monitored throughout the column study included cadmium, copper, and iron. Cadmium and copper are obvious concerns given their known presence in MSW incinerator ashes and their potentially harmful effects on various life forms even in trace concentrations (Waldbott, 1973). Iron was a parameter of interest for the same reasons previously cited for manganese; its reactive nature and dependence on redox and pH.

Concentration plots were not developed for cadmium, copper, or iron since their presence in the leachates generally fell at or below detectable limits. Trace levels of cadmium and copper in the leachates were expected given their low concentrations in the MSW combined residue. Conversely, iron was present in the residue at levels exceeding 40,000 ppm. Batch tests also failed to leach

significant concentrations of iron from the nine various unamended and amended residue matrices. The failure of iron to leach from all of residue matrices tested in both the dynamic and static leaching tests, indicates it is relatively inert and unavailable for leaching under typical environmental conditions.

Cation & Anion Equivalent Concentrations

The anions and metals selected for analysis were based on the results of previous studies. These selected constituents do not fully represent the entire spectrum of anions and metals associated with the incinerator residue, but hopefully encompass the principle cationic and anionic species. As a test, Figure 3.18 was developed comparing the summation of measured cation equivalent concentration in each respective column leachate to their corresponding anion equivalent concentration.

A comparison of the cement cation and anion equivalents plots indicate that the principle metal species released through 0.1 liquid/solid ratio were in the form of metal chlorides and sulfates. After this liquid/solid ratio, a constant difference of 30 meq/l is apparent throughout the study, thus suggesting certain significant anionic species may be present in the leachate and going unmeasured. The high relative alkalinity associated with the cement amended leachate suggests that the unmeasured anions may be



FIGURE 3.18: Comparison of Cation and Anion Equivalent Concentrations Measured in Column Effluent Leachates

carbonates, bicarbonates, and hydroxides. Past studies have also found significant phosphate and fluoride concentrations exist in leachates from MSW incinerator residues (van der Sloot et al. 1987). Further analysis for these anions may result in narrowing the difference between cation and anion equivalent concentrations and assist in identifying the speciated forms of the residue related metals.

A similar comparison of the lime cation and anion equivalents show a different relationship. From the study's beginning, anion concentrations in the lime amended columns exceeded cation concentrations by approximately 80 meq/l. This trend persisted throughout the study (L/S=0.4), suggesting that metals, other than those analyzed, may have been present.

The control leachate experienced the same increased anion versus cation equivalent concentration relationship seen in the lime amended leachate, but to a far greater degree. The average anion and cation equivalent concentrations in the residue control leachate was 617 meq/l and 292 meq/l, respectively. The average difference between the anion and cation equivalents was approximately 300 meq/l.

The anion imbalance comes from the tremendous sulfate concentrations present in the leachate. Sulfate levels alone accounted for over 86% the entire anion equivalent concentration. Therefore, most of the control leachate's

unidentified cations must be in the form of metal sulfates. Further analysis of the control and lime amended leachates for potassium, magnesium, chromium, molybdenum, and strontium may provide additional insights into cation/ anion balance and overall metal speciation (Miner et al. 1986; van der Sloot et al, 1987; Hjelmar, 1987).

Assessment of Leaching Trends Within Amended Columns

After several weeks of leaching, the amended columns were retrofitted with sampling ports, or stations, to permit leachate withdrawal at one foot intervals. Leachate extracted from the various heights within the columns were analyzed for the same environmental parameters and metal concentrations as were the effluent leachates. The results of each analysis are reported relative to the height of residue material located above each sampling station. The corresponding results measured on effluent leachates collected over the 3 - 4 day period inclusive of each station sampling event are also displayed. Direct comparisons between the discrete station and composite effluent leachates are more useful in explaining the effects of atmospheric exposure and/or the columns' sand filter on final leachate mineralogy, rather than the completion of trend data relative to six additional inches of residue. Samples were withdrawn on Days 39, 86, and 146 of the column study. These sampling days corresponded to effluent

liquid/solid ratios (L/S) of 0.11, 0.25, and 0.40, respectively.

The purpose of the height study was to identify the various trends occurring within the "black box" columns, and to quantify the reducing conditions believed to be representative of an amended MSW incinerator residue landfill.

Environmental Parameter Trends. Figure 3.19 illustrates the alkalinity trends seen throughout the length of the cement and lime amended columns. Alkalinity in the cement column followed an increasing trend with increasing residue material exposure (i.e. decreasing column height). This trend was consistent over time, as seen by comparing the alkalinity results determined on Day 86 and Day 146. The lime column exhibited a similar increasing alkalinity trend measured in the three Day 86 sampling station leachates, while a fairly constant alkalinity release was seen throughout the column on Day 146. A deviation from these trends occurred with alkalinity levels dropping sharply in the lime column effluent samples relative to levels measured in the column. This may have been due to adsorption of alkalinity contributing ions in the sand filter, and/or an increase of CaCO, precipitation due to the influence of atmospheric CO, on the effluent leachates (Snoeyink & Jenkins, 1980; Lindsay, 1979).





FIGURE 3.19: Alkalinity Trends in the Cement (a) and Lime (b) Amended Columns

The prevailing pH conditions in the cement and lime columns are presented in Figure 3.20. The cement amended leachates consistently demonstrated a constant pH level (approximately 12.0) throughout the length of the column. The pH conditions within the lime column were also consistent, with a decrease in pH typically occurring in the effluent leachates. The loss in pH at the effluent corresponds to the decline in alkalinity shown in Figure The mechanism driving the decrease in pH and 3.19b. alkalinity levels between the effluent and column height leachate samples are probably related. Additionally, slight decreases in pH were apparent between sampling events of the lime amended leachates. This supports the previously stated assumption that buffering of the acidic leaching media occurs rapidly to pH and alkalinity levels characteristic of the waste form.

The results of the redox analyses for each set of amended columns are reported in Figure 3.21. The sampling port configuration permitted the measurement of the leachate's redox almost immediately after withdrawal from the center of the column. This greatly reduced the redox variability cited as a problem in the measurement of effluent leachates, by minimizing the influence of atmospheric O_2 . As seen in Figure 3.21a, a constant reducing environment existed within the cement amended column. This trend between sampling stations was





FIGURE 3.20: pH Trends in the Cement (a) and Lime (b) Amended Columns

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FIGURE 3.21: Redox Trends in the Cement (a) and Lime (b) Amended Columns

consistent, while the overall redox environment in the cement column became more reduced over time. The increasingly reduced environment may have been related to the production of $H_{2(g)}$ by the curing reaction involving residue associated aluminum and cement alkalias, and the subsequent redox couple described in Equation 3 (Drever, 1988).

Although the lime leachate's redox values differed significantly from that of the cement leachate, the constant trend between sampling station redox levels were the same. The overall redox environment inside the lime column became more oxidized between Day 39 and Day 86, and then slightly more reduced at Day 146. This may have been a response by microorganisms associated with the combined residue to the localized shifts in ionic strength and temperature occurring in the lime column over time (Atlas & Bartha, 1987). Certainly, a more extensive evaluation of the lime amended residue's microbial activity is required before the validity of this hypotheses could be accepted.

Large differences in redox were seen between both columns' effluent leachates and the leachate extracted at the three foot stations. This was expected given the established analytical protocol and the influence of oxygen on samples exposed to the atmosphere during effluent collection.

The final environmental parameter evaluated on the sampling station leachates was conductivity. Figure 3.22 illustrate the prevailing trends in each amended column with respect to conductivity. The conductivity levels seen in the both the cement and lime amended leachates demonstrated an increasing trend with increasing residue height. This was as expected since total dissolved ion concentration will tend to increase as greater contact between the residues and leaching media occurs. Similarly, the overall conductivity in the cement and lime columns decreased with each successive sampling event. The relative loss of dissolved ions over time is best explained by the gradual decrease seen in sodium, chloride, and sulfate concentrations as a function of increasing liquid/solid ratio.

The conductivity trends cited within the columns gain added significance when considering possible large scale landfilling of similarly prepared amended MSW incinerator residues. Continual deposition of fresh residues on top of existing more aged residues should increase ionic strength of landfill leachates as they percolate through the stockpiled residues. Increasing ionic strengths will inevitably increase metal solubility and potentially increase leachate toxicity, until the landfill is ultimately capped and decommissioned.

<u>Metal Leaching Trends</u>. A discussion of metal concentration data generated during the height study is





FIGURE 3.22: Conductivity Trends in the Cement (a) and Lime (b) Amended Columns

limited to those metals which exhibited the most definitive trends. In the cement and lime amended columns, the most mobile metals were calcium, aluminum, and sodium. Additionally, the release patterns of lead and manganese were traced in the cement and lime amended leachates, respectively.

Figure 3.23a illustrates the calcium concentration trends exhibited in the cement amended leachates. A decrease in dissolved calcium was seen between leachates extracted from the 1 and 2 foot residue heights, followed by a subsequent increase in calcium concentration in the 3 foot leachate samples. This trend was repeated on both sampling events, and bares little resemblance to the environmental parameter trends previously developed. Additionally, a significant decrease in dissolved calcium concentration was evident in the effluent leachates compared with sampling station leachates. Reasons for this decrease may relate to the influence of atmospheric carbon dioxide on calcium carbonate solubility, or the possible adsorption of calcium to silicate surfaces of the sand filter media (Lindsay, 1979; EPRI, 1984).

The release of calcium within the lime column followed a decreasing trend with increasing residue contact, as illustrated in Figure 3.23b. Calcium appeared to be most mobile at the three foot level as evidenced by the results of Day 86 and 146 sampling events. The calcium and



FIGURE 3.23: Calcium Concentrations Measured Within the Cement (a) and Lime (b) Amended Columns

alkalinity trends associated with the lime column suggests that calcium is removed from solution and deposited throughout the column as $CaCO_3$ precipitate. Close agreement between calculated and measured effluent calcium concentrations suggests that the predominant form of soluble calcium is $CaSO_4$ (gypsum) (Lindsay, 1979).

The amphoteric nature of aluminum is clearly demonstrated by comparing the cement and lime amended leachate samples illustrated in Figure 3.24. The relatively high concentrations of aluminum in the cement leachates corresponded to measured pH levels (approximately 12.2), which approached maximum aluminum solubility. Aluminum mobility increased in each successive station leachate, culminating in a major rise in dissolved aluminum at the effluent. This major jump in soluble aluminum is not the product of a significant pH increase, but may be a function of desorption of hydrolyzed forms of aluminum from the sand filter surfaces under high pH conditions (EPRI, 1984).

In Figure 3.24b, the first indications of aluminum release from lime amended residue was seen in the station leachate samples withdrawn on Day 86 and 146. Aluminum concentrations were far less than those measured in the cement columns as expected given the differences in pH. Similarly, aluminum was less mobile in the Day 146 leachates than in the Day 86 leachates. The corresponding pH levels for the Days 86 & 146 station leachate averaged 9.13 and





FIGURE 3.24: Aluminum Concentrations Measured Within the Cement (a) and Lime (b) Amended Columns

9.08, respectively. This slight pH difference reflected a concomitant disparity in aluminum concentrations. The difference in aluminum concentrations between the lime column effluent and sampling station leachates are similarly explained by measured pH differences.

Sodium concentrations and release trends associated with the cement and lime amended leachates were nearly identical, as shown in Figure 3.25. The trends reflect sodium's conservative nature, demonstrating increased mobility with increased residue contact, as well as decreased concentrations over time. These similarities suggest that the MSW combined residue must be the major source of the sodium dissolved in the leachates.

Lead mobility appeared suddenly in the Day 146 station and effluent cement amended leachates, as shown in Figure 3.26. The release of lead was not predicated on a major shift in leachate pH conditions, therefore suggesting that its mobility may have been more a function of desorption than solubility. Past studies have suggested that lead participates in surface complexation reactions with iron, manganese, and aluminum oxides, as well as organic matter and silica (Barrow et al., 1981; Hohl & Stumm, 1976; Davis & Leckie, 1978; Schindler et al.,1976). Dissolution of various lead minerals (e.g. PbSO4) may have been occurring throughout the column study due to the high prevailing pH conditions, but removed from solution by the subsequent





FIGURE 3.25: Sodium Concentrations Measured Within the Cement (a) and Lime (b) Amended Columns



FIGURE 3.26: Lead Concentrations Measured Within the Cement Amended Columns



FIGURE 3.27: Manganese Concentrations Measured Within the Lime Amended Columns

adsorption onto the aforementioned surfaces. Adsorption of aqueous lead persisted until a threshold liquid/solid ratio was achieved signaling the exhaustion of available surface sites and the resulting detection of lead in the cement leachates. This theory makes many unsubstantiated assumptions that certainly require verification and more detailed investigation prior to acceptance.

Undetectable manganese levels prevailed in lime station leachates, while significant manganese releases were measured in the column's effluent samples, as shown in Figure 3.27. The release of manganese only in the effluent leachate suggests that the sand filter may have been involved in manganese mobility. A possible explanation relies on the results of previous studies which cite Mn(II)'s capacity to adsorb to calcium carbonate, iron oxide, and manganese oxide surfaces (McBride, 1979; Takematsu, 1979). The environmental conditions measured within the station leachates favor the existence of Mn(II) compounds, therefore suggesting that manganese may have been bound to these surfaces and effectively immobilized (EPRI, 1984; Lindsay, 1979). Manganese may have subsequently released from the solid through an ion exchange process with transition metals possibly associated with the sand filter media (Donner et al., 1982). Additional study characterizing both the mineral phases within the lime columns, as well as the sand filter's elemental composition

would certainly assist in validating/ invalidating this hypothesis.

Batch Study Results

Individual batch experiments were performed on each amended MSW incinerator residue matrix to assess their potential to leach heavy metal under a controlled aggressive environment. Batch extraction studies offered an effective means to simulate long term leaching scenarios in relatively short time periods. Typically, batch studies are considered simpler to perform, less costly, and more reproducible than column leaching studies (Jackson et al, 1984). The batch leaching experiments conducted on the residues provided a means to qualitatively assess characteristic leaching profiles of cement, lime, and asphalt amended residues, as well as identified relative metal release trends associated with the various residue/amendment mixtures. Additionally, the batch experiments permitted focused evaluation of trends cited previously during the column study.

Residue Buffering Capacity

Most metals become more mobile under increasingly acidic conditions. Therefore, the buffering capacity provided by the solidification/ stabilization agent to the amended waste form represents a viable shield against metals release. Although each amended and unamended residue's absolute buffering capacity was not determined, the batch extraction experiment on pulverized residues provided a relative assessment of matrix buffering capacity through the precise measurement of titrant volume required to achieve and maintain a pH 6 condition. Direct comparison of the various residue matrices was based on the resulting titrant equivalent concentrations.

In Figure 3.28, the calculated titrant equivalent concentrations required to sustain pH 6 conditions for each amended material are featured as realistic environmental leaching periods. These leaching periods (in thousand years) represent the residues' minimum exposure time to precipitation typical of the New England area. Specifically, the leaching period determination assumes the waste will see an average rainfall of 53 inches per year, with a total acidity of 210 ueg/l (Pierson & Chang, 1986). All precipitation is assumed to infiltrate a one cubic foot volume of pulverized waste material compacted to a uniform packing density specific to the residue. The packing density achieved in the cement, lime, and control residue columns were used as each material's characteristic packing density value in the leaching period calculations. A similar determination of the characteristic packing density was also accomplished for the asphalt amended residues. The leaching period determinations conducted on pulverized



FIGURE 3.28: Environmental Leaching Period Equivalent to Sustained pH 6 Conditions for Each Amended Residue Matrix

amended residues are relative estimates that are conservative in nature. Leaching of monolith materials would most likely require even greater leaching periods to achieve metals concentration levels similar to those measured during the batch extraction tests.

The trends illustrated in Figure 3.28 indicate that the addition of cement and lime amendments to MSW incinerator combined ash increased its overall buffering capacity and ability to resist acidic attack. The buffering capacity relationship of the residues corresponded with stoichiometric addition of cement and lime amendments. Conversely, the increased addition of asphalt appeared to have an inverse effect on matrix buffering capacity. In general, all asphalt amended residues demonstrated a reduced capacity to buffer against acidic conditions, suggesting that the amendment imparts a degree of acidity to the combined residue. The significance of this observation may be irrelevant if the asphaltic residues' leaching year values determined using this procedures (i.e. 7,000 - 16,000 years) are realistic.

Metal Leaching Profiles

The leaching of ten metals from pulverized unamended and amended residues was measured at discrete time intervals under controlled pH conditions of 10, 8, and 6. Metal release profiles were determined respective to the specific

residue matrix and pH condition. These individual kinetic trends were combined graphically to portray and contrast the response of each amended residue matrix to an environment which becomes progressively acidic. Additionally, bar graphs were created which permit direct comparison of metal dissolution rates from various residue materials under each given pH condition. The rate of dissolution, or release constant, of a specific metal was taken as the slope of the correlation between leaching results and time.

The significance of the calculated release constant is labeled above each respective bar plot. The confidence level listed was based on the statistical evaluation of the correlation coefficient, r, determined for each correlation equation (Lipson & Sheth, 1973). Bar plots without confidence level labels represent release constants that were significant at less than 90% confidence. Since the bar plots are used solely to relate trends, not absolute data, the inclusion of results determined at less than 90% confidence is justified. The use of these release constants permits the direct comparison of residue matrices that differ dramatically in metal concentration range, as well as providing general estimates of future matrix behavior.

<u>Nickel</u>. The effects of increased acidity and time on the release of nickel from individual residue matrices are depicted in Figure 3.29. Direct comparison of trends shown in the figure are possible since nickel was measured in



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similar concentrations in each of the tested leachates. Nickel concentrations measured in the leachates drawn at pH 10 and 8 were at or below detectable levels, while pH 6 leachates exhibited only trace levels of nickel.

Nickel was relatively immobile until subjected to a pH environment of 6. At this pH, all residues (except the 26% asphalt) leached increasing nickel concentrations over the sampling interval. These increasing trends appeared to continue past the selected time interval, indicating that nickel release from the residues failed to achieve relative equilibrium during the batch experiment. This was expected since similar batch leaching tests performed on the residue control material for 24 hours also failed to establish equilibrium with respect to nickel dissolution (see Appendix).

A comparison of nickel release trends among the cement amended matrices at pH 6 reveals an inconsistency. The 17% cement matrix released more nickel than the control, 33% cement, and 11% cement residues, respectively. Similar conflicting relationships are seen for the asphalt and lime amended residues at pH 6. The inability to establish either increasing, or decreasing trends suggests the absence of a relationship between percent amendment addition and nickel solubility, or the influence of experimental error on the results. The inclination is to accept the latter explanation since variability in the designed amendment/ residue mix ratio, or in the combined residue itself, could have a major effect on the results of the leaching experiment, especially considering the small nickel concentration ranges and replicate analyses involved.

In Figure 3.30, a clearer definition of nickel release trends is apparent through illustration of each amendment's nickel release rate constants. As previously described, no appreciable nickel dissolution occurred prior to pH 6 leachate testing. The highest release constant was associated with the 17% cement residue which exceeded that of the control by approximately 21%. All other amended residues maintained lower release constants than that of the control suggesting possible solidification/ stabilization of nickel by the respective amendment.

Zinc. Zinc release trends for each residue matrix bore a strong resemblance to the respective nickel release trends previously described. Figure 3.31 illustrates these trends that differ from nickel only in the magnitude of soluble metal measured in the leachates. Zinc mobility was not observed in any residue leachates prior to pH 6, where its release demonstrated increasing kinetic trends in all tested materials. As with nickel, the dissolution of zinc from the residues failed to reach equilibrium within the batch experiment's 4 hour sampling interval; nor did zinc approach equilibrium in the extended 24 hour leaching test performed on the unamended control residue (Appendix).



FIGURE 3.30: Nickel Release Constants Determined for Each Amended Residue Matrix Under pH 10, 8, and 6 Conditions




The leaching trend of zinc from the cement amended residues under pH 6 conditions demonstrated the same inconsistency as nickel. Variability with respect to zinc concentrations in the combined MSW incinerator residue is suspected as the primary reason for the inconsistent release trends. The 10% and 5% lime residues zinc release trends appeared to differ from that observed for the residue control, while little difference was cited between each lime amended matrix. Similarly, the 18% and 22% asphalt matrices showed little difference in their respective zinc releases, while an increased release of zinc was seen for the 26% asphalt residue. Given the magnitude of this difference, this higher release most likely represents incomplete coverage or encapsulation of the residue by the asphalt binder.

The zinc release constants and associated confidence levels for each residue matrix are shown in Figure 3.32. The improvement in the significance of the release constants accurately reflects the almost linear zinc release pattern inherent in all the residue leachates tested. The small relative differences between the various release constants suggests that zinc mobility under controlled acidic conditions is independent of amendment type, and primarily a function of the aggressiveness of the surrounding environment.



FIGURE 3.32: Zinc Release Constants Determined for Each Amended Residue Matrix Under pH 10, 8, and 6 Conditions

<u>Cadmium</u>. Overall levels of cadmium in the residue leachates were among the lowest of all detectable metals. Figure 3.33 illustrates the cadmium levels for each leachate tested, demonstrating release trends similar to zinc in statistical significance, and characteristic concentrations more analogous to those of nickel. Cadmium became mobile only under the most acidic conditions tested, following an increasing release trend in all residue leachates. Neither the 4 or 24 hour batch leaching tests provided sufficient time to reach an equilibrium release condition for cadmium, indicating that either longer leaching periods or harsher leaching conditions may be required.

Higher concentrations of cadmium were present in the control leachate than all other amended leachates tested at the pH 6 condition. This suggests that amending MSW incinerator residues decreases cadmium mobility. The relative release constants calculated for each residue matrix also supports this observation as shown in Figure 3.34. The levels of significance for the cadmium release constants reflect the linearity of the batch leaching results. This was unexpected given the trace concentrations of cadmium measured in all the residue leachates. Cadmium immobilization appears to be enhanced by the addition of an amendment, but unaffected by amendment type or quantity. Future study is required to determine possible reasons for this occurrence.



Cadmium Release Profiles for Each Cement, Lime, and Asphalt Amended Residue Matrix FIGURE 3.33:



FIGURE 3.34: Cadmium Release Constants Determined for Each Amended Residue Matrix Under pH 10, 8, and 6 Conditions

Copper. The leaching of copper from individual residues also occurred only under the most acidic testing conditions. Figure 3.35 portrays the relative copper release trends associated with each residue matrix as a function of pH and time. Overall leachable concentrations of copper were double that of cadmium, but more than an order of magnitude less than measured zinc concentrations. Release of copper during the pH 6 batch leaching experiment appeared to approach relative equilibrium in the cement and lime amended leachates, but not in the asphalt amended and residue control leachates. This suggests solidification/ stabilization of copper by the cement and lime amendments may effectively occur even under conditions that promote copper mobility from unamended waste forms.

Similar observations concerning cement and lime's solidification\ stabilization potential may be drawn by comparing their respective copper release constants shown in Figure 3.36. The copper release constants for lime, cement, and to a lesser extent asphalt amended materials were much less than that of the control. Additionally, the trend toward a slight decreased mobility with increased amendment addition appears to exist for both the lime and asphalt residues. Conversely, the cement residues exhibited neither a decreasing nor increasing copper mobility trend, maintaining the greatest release constant for the 17% cement



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FIGURE 3.36: Copper Release Constants Determined for Each Amended Residue Matrix Under pH 10, 8, and 6 Conditions

matrix. Non-homogeneity of the combined residue was again suspected as the primary cause for the trend inconsistency.

Aluminum. Aluminum demonstrated an inverse release profile maintaining its greatest mobility under pH 10 conditions, while failing to release detectable concentrations at either pH 8 or 6. This was expected given aluminum's amphoteric nature and the results from the column leaching of similarly amended materials. Figure 3.37 also shows a relative equilibrium condition existing in the cement amended matrices with respect to aluminum release. Higher initial aluminum mobility characterizes the cement matrices at pH 10 compared with other similarly treated amended residues. The higher mobility was most likely caused by experimental variability in achieving and maintaining a uniform pH 10 condition in the more buffered cement amended batch reactors. Nevertheless, the higher aluminum mobility seen in the control leachate suggests that aluminum may be effectively solidified or stabilized in the various amendments. The precipitation of aluminum at more neutral pHs is apparent in the control leachate as seen by its sharp decrease in aluminum concentration measured during the first 30 minutes of pH 8 batch testing.

The relative aluminum release constants for the various residues are shown in Figure 3.38. The magnitude of these constants at pH 10 far exceeds all previously described metal release constants determined under any pH condition







FIGURE 3.38: Aluminum Release Constants Determined for Each Amended Residue Matrix Under pH 10, 8, and 6 Conditions

tested. This may prove to be significant in light of the increased concern over aluminum toxicity and its potential link to various neurological disorders (Waldbott, 1973). Trends among matrices of a particular amendments were largely inconsistent, again reflecting possible nonhomogeneity of combined residue used in the amending process.

Calcium. Calcium was selected as an analytical parameter due to the significant concentrations known to exist in both the MSW incinerator residue and the cement and lime amendments. High levels of calcium were measured in all residue leachates extracted during every pH leaching condition, as expected. In general, these levels far exceeded the measured concentrations determined for any other analyte during the batch study. Figure 3.39 illustrates the magnitude of dissolved calcium as well as its relative release trends associated with each respective residue matrix. Calcium demonstrated increased solubility with decreasing pH in all residues. This confirmed previous leaching trends identified during the column study and is consistent with established calcium carbonate behavior found in the literature (Lindsay, 1979; Stumm & Morgan, 1981).

Near equilibrium conditions with respect to calcium release were seen for all the residue matrices at the pH 10 condition. The pH 10 calcium concentrations measured in all but the 33% and 17% cement leachates were characteristic of





levels seen in the control. More distinct trends began to evolve as the leaching environment became more acidic. Nonequilibrium trends with respect to calcium release were seen for both the cement and asphalt residues at pH 8 and 6, while relative equilibrium conditions prevailed in the lime matrices until subjected to pH 6 leaching. Release trends showed a decreased calcium mobility in the asphalt amended residues compared to the control during both the pH 8 and 6 leaching experiments, while cement residues developed increased calcium mobility relative to the control over the same pH conditions. Increased calcium mobility in the lime amended leachates relative to the control was not apparent until pH 6 leaching.

The strong correlation between decreasing pH and increasing calcium solubility is clearly shown for all residues tested in Figure 3.40. Equally apparent is the correlation between increased amendment addition and calcium mobility for both the cement and lime residue leachates, while the opposite relationship is seen for the asphalt amended residues. Effective immobilization of calcium from MSW incinerator residues appears to occur in the asphalt amended residues, while cement and lime amendments exacerbate calcium release.

<u>Manganese</u>. Manganese paralleled zinc with respect to release trends and overall dissolved concentrations measured in the leachates. No appreciable manganese release was seen



FIGURE 3.40: Calcium Release Constants Determined for Each Amended Residue Matrix Under pH 10, 8, and 6 Conditions

prior to pH 6 batch testing, as shown in Figure 3.41. This conflicts with previous leaching results generated during the 5% lime and control column studies, which showed significant manganese release at pHs as high as 9. The major difference between redox environment represents the most likely explanation for this inconsistency. Past studies have shown that manganous ions may be removed from solution and incorporated into solid phases with manganese oxides under highly oxidizing conditions (McKenzie, 1980; Posselt et al., 1978). Batch leaching was conducted at mixing speeds exceeding 400 RPMs, which unavoidably induced a highly oxidized state within the reactor. Redox measured in both the lime and control column leachates were not as oxidized.

The trends developed under pH 6 leaching show all residues failing to achieve relative equilibrium with respect to manganese release within the 4 hour sampling interval. Leaching of the control residue over a 24 hour sampling period also demonstrated the same non-equilibrium condition. Future leaching test of similar design may need to extent over several days or weeks to eventually leach all available manganese fractions.

The manganese release constants associated with each individual residue matrix differed only slightly, as shown in Figure 3.42. This suggests that little difference between the amendment agents, or the relative percentage of







FIGURE 3.42: Manganese Release Constants Determined for Each Amended Residue Matrix Under pH 10, 8, and 6 Conditions

those agents, exists with respect to immobilizing manganese from MSW incinerator residues. The relative significance of the rate constants also indicates the linearity of manganese release over the given leaching conditions.

Sodium. Relative equilibrium with respect to sodium release was apparent for all tested matrices at all pH conditions. Figure 3.43 illustrates the sodium release trends for each residue matrix. Sodium mobility appears to occur almost immediately upon contact with the leaching media as evidenced by the high sodium concentrations measured in each of the residue's initial pH 10 leachate samples. In fact, all residues (except 5% lime) leached over 90% of their total pH 6 leachable sodium within the first 60 minutes of the pH 10 batch experiment.

The 5% lime and, to a lesser extent, control residues showed increased sodium mobility with a corresponding decrease in pH from 10 to 8. Additionally, much lower sodium concentrations were measured in the control leachate withdrawn under the pH 8 extended batch leaching experiment than were measured during the shorter 4 hour batch experiment (Appendix). These trends illustrate the variability inherent in the residue with respect to sodium content and support published literature concerning sodium's non-reactive nature. Insights in the nature or cause of the increased sodium mobility in the control and 5% lime leachate requires replicate testing to determine if the





release is more a function of ash heterogeneity rather than increased acidity.

Sodium rate constants were not calculated due to the level or horizontal sodium release profiles exhibited by all the residues leachates.

Lead & Iron. All residue leachates generated during the batch study failed to possess concentrations of lead and iron in detectable levels. The release of lead from the residues during batch leaching was of particular interest given its acute toxicity to humans and animals and its sudden mobility in the column study's 17% cement leachate. The average pH and E, conditions prevailing during lead release in the column study was 11.6 and -40 mv, respectively. Conditions during batch leaching of the residue materials never approached these levels, maintaining pH ranges between 10 and 6, as well as highly oxidized conditions within the batch reactor. Lead is known to exist in various mineral phases under these given environmental conditions, favoring the formation of PbCO, at pH 10 and 8, while pH 6 conditions favor the more stable PbSO₄ (Lindsay, 1979).

Iron failed to release in significant concentrations from the residues in both the column and batch studies. The pH and E_h conditions predominating during the column and batch leaching experiments ranged from 6 to 12.7, and -300 mv to greater than 400 mv, respectively. The attenuation of iron over such wide ranging environmental parameters suggests its presence in the residues as $Fe(OH)_3$ and Fe_2O_3 minerals (Lindsay, 1979; Drever, 1988).

Comparative Metal Concentrations

A summary of batch study results emphasizes the vast disparities that existed between the metal concentration ranges leached from various amended and unamended MSW incinerator residues. Figures 3.44 & 3.45 illustrate the peak concentrations of all metals measured in the residue leachates at pHs 10 and 6, respectively. Due to the enormous scale required to display aqueous calcium and sodium levels, Figures 3.46 & 3.47 were developed which show the relationships between the concentrations ranges of the remaining eight metals over the same pH conditions. Metals are arranged along the graphs' abscissa in order of decreasing concentration (left to right).

Calcium and sodium were the predominant cations leached from every residue under all pH conditions evaluated during the batch study. A comparison of Figures 3.44 & 3.45 showed dramatic increases in calcium concentrations as the environment became more acidic, while sodium levels remained relatively constant reflecting sodium's conservative nature.

Considering metals other than calcium and sodium revealed that only aluminum was leached in significant concentrations at pH 10, as shown in Figure 3.46. The



FIGURE 3.44: Peak Concentrations Exhibited by Amended and Unamended Residues Under pH 10 Conditions Inclusive of Calcium and Sodium



FIGURE 3.45: Peak Concentrations Exhibited by Amended and Unamended Residues Under pH 6 Conditions Inclusive of Calcium and Sodium



FIGURE 3.46: Peak Concentrations Exhibited by Amended and Unamended Residues Under pH 10 Conditions Exclusive of Calcium and Sodium



FIGURE 3.47: Peak Concentrations Exhibited by Amended and Unamended Residues Under pH 10 Conditions Exclusive of Calcium and Sodium

relative order of the most mobile metals changed as the pH of the leaching environment decreased. Figure 3.47 depicts the adjusted order with zinc and manganese gaining in overall mobility under the pH 6 batch leaching conditions.

Presentation of batch leaching results in this manner allows direct comparisons of metal concentration levels leached from the various amended/unamended residues and permits the qualitative assessment of each metal's true environmental significance based on established toxicity data.

Correlation of Batch and Column Study Results

The results from the unamended pH 8 batch leaching experiment and the control columns were used to assess the overall utility of the batch design in modeling long term leaching characteristic of MSW incinerator residues. Comparisons of the metal concentration data measured in the batch and column leachates were not expected to yield statistically significant findings given the differences in pH, redox, and particle size between the respective leaching studies. The comparisons were more focused on the batch experiment's ability to duplicate leaching trends established in the more expensive and time consuming column studies.

Figure 3.48 illustrates the relationship between metal leaching trends measured in the combined residue (control) leachates generated under the batch experiment's controlled pH 8, and column study's average pH 7.79 conditions. In general, the batch results overestimated metal leachability as measured in the control columns, but appeared to exhibit similar release trends. The most pronounced deviation from this pattern was seen with respect to manganese release. Manganese was undetectable in batch leachates while significant concentration were measured in the control column samples. Reasons for this deviation may be related to manganese's propensity to incorporate into solid phases with manganese oxides under highly oxidizing conditions similar to those prevailing in the batch reactor (McKenzie, 1980; Posselt et al., 1978). Aluminum concentrations were also undetectable in the batch experiment while demonstrating limited mobility in column leachates. The difference was small and may be more due to analytical variability than differences in solution chemistry.

The batch leaching experiment appeared to adequately predict predominant cation release trends demonstrated in the column leaching study. The generation of the data was accomplished at a significant time and cost savings. Similarly designed leaching experiments would be useful in preliminarily assessing the characteristic leaching trends associated with other residue materials, and possibly



FIGURE 3.48: Comparison of Metal Leaching Trends Derived from the pH 8 Unamended Batch and Control Column Leaching Experiments

provide information beneficial in electing to pursue more complex and expensive column leaching experiments. ,

CHAPTER IV

CONCLUSIONS

The performance of cement, lime, and asphalt amended municipal solid waste incinerator residues was evaluated from numerous perspectives. Conclusions are grouped according to the major activities contained within the scope of this research. The activities include material strength properties, column leachate parameters, and batch leaching trends.

The following list represents the principal conclusions concerning the material properties of the amended MSW incinerator residues:

- Cement and lime amended specimens demonstrated increasing compressive strength with increasing amendment addition
- Residue and lime reacted pozzolonically as seen by the specimens' increased compressive strength with increased curing time
- Compressive strength and Marshall stability increased to a maximum at 23% asphalt addition. 2000 lb Marshall stability was achieved at 23% asphalt content, and flow of 23

- Volume of asphalt addition was insufficient to fully

encapsulate the combined incinerator residue due to the the residue's vast pore structure

Leachates generated from crushed 17% cement amended, 5% lime amended, and residue control columns were periodically analyzed for alkalinity, pH, redox, and conductivity, as well as calcium, sodium, lead, aluminum, manganese, zinc, nickel, cadmium, copper, and iron concentrations. The results of the analyses indicated:

- Calcium and sodium were the predominant cations in the leachates
- Sulfate was the predominant anion
- Iron, cadmium, and copper leached in undetectable levels in all leachates
- Nickel and zinc were present in the leachates in only trace concentrations
- Amending the residue with 17% cement reduced the ionic strength and redox, while increasing the buffering capacity and pH of the resulting leachate. The cement addition attenuated calcium, manganese, and sodium mobility and exacerbated the release of lead and aluminum from the combined residue
- The 5% lime addition decreased the buffering capacity pH, ionic strength, and redox of the residue leachates.

Manganese and sodium experienced decreased mobility in the lime amended leachates, while all other metals leached in concentrations characteristic of the residue leachates

Batch leaching of eight different amended residue matrices was conducted to distinguish metal release trends and assess the test's capability to simulate realistic metal leachability from residue related materials as determined by more expensive and time consuming experiments. The results of the analyses yielded the following conclusions:

- Ability of finely ground (>0.3 mm) cement and lime amended residues to buffer against acidic attack increased with increasing amendment addition.
- Buffering capacity of similarly prepared asphalt amended residues decreased with increased asphalt addition
- Calcium, sodium, copper, cadmium, iron, nickel, zinc, and manganese exhibited increased mobility with decreased pH in all amended and unamended residue leachates
- Lead failed to leach in detectable levels from residues under controlled pH 10, 8, and 6 conditions
- Aluminum release from all residue materials was

significant only under pH 10 leaching conditions

- Amending the residue with cement decreased the overall mobility and release rate of cadmium, manganese, and aluminum
- Lime amended residues demonstrated decreased mobility and release rates for zinc, cadmium, copper, aluminum, and manganese
- Asphalt amended residues demonstrated decreased mobility and release rates for manganese, calcium, aluminum, and cadmium
- Varying amendment addition had little effect on metal concentrations and leaching trends measured in the respective leachates. Calcium was the only exception demonstrating increased release with increasing cement and lime amendment, and decreased release with increasing asphalt content.
- Batch experiment correctly predicted predominant cations (Ca, Na) and relative release trends as determined in column leaching studies of the control residue. In general, estimates of metal leachability were far greater in the batch leachates than in the column leachates. Batch experiments underestimated manganese, and to a lesser extent aluminum, releases relative to the column study results

Overall, the average precision and accuracy of the various analyses were acceptable for the purposes of this research. Most metals increased mobility as the pH of the leaching environment drifted from neutral conditions. The results are not compelling, but appear to favor lime as a MSW incinerator residue stabilization agent over cement and asphalt due primarily to its ability to buffer the surrounding environment at or near a pH of 7. More focused evaluation of all the MSW incinerator residue solidification/ stabilization agents is required.

CHAPTER V

RECOMMENDATIONS

Recommendations concerning the focus and conduct of future research relating to solidification/ stabilization of municipal solid waste incinerator residues are similarly classified according to major activities. Assessments of the material strength properties of cement, lime, and asphalt amended residue should:

- Increase specimen replication to enable greater statistical evaluation of results. This is especially important for asphalt amended specimens given the difficultly associated with fabricating a homogeneous mix design
- Extend curing time and subsequent testing of lime specimens to further evaluate the limits of its pozzolonic activity
- Incorporate conventional aggregate into residue mix designs to increase structural integrity, ease fabrication costs, and enhance potential for reutilization as a construction material
- Consider solidifying/ stabilizing water saturated MSW incinerator residues using asphalt emulsion techniques. Saturating residue pores prior to emulsification may reduce the volume of asphalt binder absorbed and

promote more complete coverage of the waste material.

- Always paraffin coat asphalt amended specimens during Marshall Mix testing
- Add required water via a fine spray nozzle during batching of cement and lime amended residues to ensure equal distribution

The use of column studies to assess leaching characteristics of amended and unamended MSW incinerator residues should:

- Continue the sampling and analyses of station leachates as an integral part of the column study. Results generated by the height studies should provide insights into landfill conditions and identify trends that may develop within an actual landfill
- Replace the sand filter media with a thin, chemically inert geotech fabric. Additionally, install the fabric over the piezometer ports to retain the residue during station sampling
- Use clear acrylic columns that provide visible inspection of column residue during leaching, thereby reducing the possibility of channelling
- Establish a constant controlled temperature in which to operate the column during leaching
- Install air tight effluent collection bottles and connections to minimize atmospheric O_2 and CO_2 influence on leachate composition
- Maintain residue and amendment control columns to better assess the contributions made by the amendment material to each individual leachate parameter

The batch experimental design simulated long term leaching of numerous residues under progressively acidic conditions. The test differed significantly from many standard batch experiments currently in use by regulatory agencies. Regardless, the batch design provided a means to compare metal mobility from wastes containing various combined ash/amendment blends, as well as yielded insights into the kinetic leaching profiles of specified metals under controlled conditions. Future studies adopting the same batch leaching design should:

 Correlate results against leaching data generated from the same residue material using various standard tests to include EP Toxicity, TCLP I, and TCLP II.
 Additionally, investigate the effects of changing leaching media and residue material when adjusting pH control to the next lower level

- Perform batch experiments at the same L/S and mixing

regime, but controlled at the average ambient pH measured in the column study. This should enable a more equitable assessment of the design's capability to adequately predict realistic leaching characteristics using small bench scale equipment

- Expand the experimental parameters to include wider pH ranges, longer leaching periods, and optional redox control measures (e.g. glove box w/nitrogen).

The follow suggestions are general in nature and concern analytical techniques and leaching protocols that may be beneficial in future study of MSW incinerator residues:

- Expand the "fingerprint" of the leachate to include analyses for silica and potassium. The former would provide insights into the overall sorption capacity inherent in the waste, while the latter figures to be important in understanding possible exchange reactions with aluminum and calcium which appeared to be significant in the cement column leachates. Consider analyses for fluoride, carbonates, magnesium, molybdenum, strontium, and chromium in an effort to narrow the imbalance between the measured anions and cations equivalent concentrations

- Perform periodic platinum probe redox measurements of column leachates collecting outside and inside an oxygen purged environment. The comparison should quantify the influx of O_2 due to atmospheric exposure and help assess the impact of the influx on metal speciation
- Consider direct analyses of redox sensitive species such as oxygen, hydrogen sulfide, and methane to determine the redox environment of a leaching system. The comparison of this data with probe measured data, may serve to "calibrate" the probe or signal when the probe may be malfunctioning
- Analyze leachates for both dissolved and total metal content. This will permit the calculation of available/ nonavailable metal fractions and lend insights into possible metal speciations
- Increase fly ash percentage in the residue or spike residues with known metal concentration to improve their detectability in the residue leachates and assist in defining release trends
- Use controlled experiments to focus on the more important or surprising trends identified in this research. Specifically, the dramatic lead release in the cement column leachates, and the apparent anomaly

between the control and lime amended leachate's measured alkalinity levels

- Perform x-ray diffraction analysis of residue, amended residues, and sand filter media to identify predominating solid phases
- Incorporate leaching of amended residue monoliths to enable better assessment of its true environmental threat. Leaching studies using crushed amended residues yield data based on conservative, worse-case, and arguably unrealistic conditions. Such studies ignore the major advantage inherent in many solidification/ stabilization agents; structural integrity
- Confirm batch and column leaching study results with larger scale pilot studies. This should provide more realistic leaching data, as well as furnish essential information on the validity of smaller, less expensive, laboratory scale leaching experiments

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IRRLE R.12: Average Netal Concentration Data for Control Column

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IFBLE A.13: Sumetion of Cation & Anion Equivalent Concentrations

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	S	Residen	9.007	9.9.9	6.015	109.6	0.00	0.820	6.639	9-9-6	609-9	0.01	9,003	9.106	9.124	0.148	0.153	0.167	6.12	0.198	9.282	0.223	0.23	996	0.10		BX B
	Sunc-)		197.28	228.888	220.022	234.121	22	228.191	197.47	166. 131	126.091																
	S	1	0.011	0.017	0.034	0,067	9.119	9.1%	0.267	0. W	0,30																
	Sunces	ŝ		89.29	50.2	299.01	198.621	104.80K	166.388	98. A	106.562	600°.701	10.636	141.071	119.682	111.392	99.219	8 9									
	ŝ	11	0.011	6.017	0.623	9-63-	9.97	9.067	0.006	0.110	0.141	0.100	<b>9</b> .1%	2,0	9.267	<b>10</b>	9.40	8.335									
	(-) HIG	ŝ	959.26	116.61	103.000	182.58	101.6M	969-92	69.991	<b>28</b> .66	8.8	67.49	₽.0°	<b>4</b> , <b>9</b> , <b>6</b>													
	S	j	0.000	8.815	0.031	999.0	0.100	0.109	0.260	0.53	9.388	6-432	0.571	9.623													
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FIGURE A.h: Copper Release Profile During Extended Run



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