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THE MODELING OF CONTAMINANT FLOW DURING PROPOSED TREATMENT OF U.S. DEPARTMENT OF ENERGY LOW-LEVEL RADIOACTIVE MIXED WASTES*

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THE MODELING OF CONTAMINANT FLOW DURING PROPOSED TREATMENT OF U.S. DEPARTMENT OF ENERGY LOW-LEVEL RADIOACTIVE MIXED WASTES*

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ABSTRACT

Estimations of waste materials throughput and the potential radiological and chemical releases resulting from the proposed treatment of U.S. Department of Energy (DOE) low-level mixed wastes (LLMWs) were used to support analyses of risks and costs associated with various waste management alternatives outlined in the Office of Environmental Management Programmatic Environmental Impact Statement (EM PEIS). The modeling of material flow and contaminant releases through a consolidated waste management flowchart was performed by the WASTE_MGMT computational model developed by Argonne National Laboratory. This paper (1) briefly describes the process used to model estimated material and contaminant flow through the proposed treatment scenarios for the EM PEIS, (2) discusses the key site- and/or waste-stream-dependent factors involved in the determination of radiological and chemical emissions, and (3) explains the assumptions used to integrate the available LLMW database with the computational model.

INTRODUCTION

The November 1994 internal draft of the U.S. Department of Energy (DOE) Office of Environmental Management Programmatic Environmental Impact Statement (EM PEIS) addresses the environmental and health risks and the costs associated with managing DOE radioactive and hazardous wastes. An important part of this effort is the identification and quantification of the risks and costs resulting from the treatment, storage, and disposal (TSD) of DOE low-level radioactive mixed wastes (LLMWs). LLMW is considered both hazardous under the guidelines of the Resource Conservation and Recovery Act (RCRA) (Title 40, Part 261 of the *Code of Federal Regulations* [CFR]) and meets the definition of low-level waste (LLW) under the 1954 Atomic Energy Act (codified under 10 CFR, Part 61).

To support the EM PEIS effort to determine the risks and costs of LLMW management, Argonne National Laboratory (ANL) estimated source terms for chemical and radiological contaminant emissions and for the waste material flow during the proposed processing of LLMW. The source terms were estimated by applying waste input data derived from various DOE LLMW databases to a computational model that tracks the flow of materials and contaminants through the proposed waste-processing facilities. The model does not track contaminant emissions from waste disposal facilities.

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The source term results from the model have been utilized in the overall EM PEIS program to (1) calculate chemical and radiological risks to site workers and the general public, (2) estimate waste-processing facility costs and resource requirements, (3) project the risks associated with transportation of LLMW and disposal products, and (4) provide comparative estimates of LLMW management activities across sites under various alternatives.

This paper briefly (1) describes the model used to estimate material and contaminant flow under the various proposed treatment scenarios for the EM PEIS, (2) discusses the key site- and/or waste-stream-dependent factors involved in the determination of radiological and chemical emissions, and (3) explains the assumptions used to integrate the available LLMW database with the computational model.

METHODOLOGY

The modeling of proposed LLMW treatment is based on a consolidated waste management flowchart (CWMF) developed by the Mixed Waste Treatment Project (MWTP) (1). The flowchart consists of a series of waste-processing modules common to all sites; the modules are designed to process the major LLMW streams identified in the 1994 Mixed Waste Inventory Report (MWIR-2) (2). Each module delineates one step in waste processing and may include one or more distinct process technologies. The complete treatment sequence for each waste type is called the waste "treatment train." The tracking of material and contaminant flow through the waste-processing modules identified in the CWMF is accomplished by the WASTE_MGMT computational model, an analytical tool that quantifies the throughput of LLMW and estimates the emissions of radionuclides and hazardous chemicals during the operation of proposed waste-processing facilities (3).

The overall modeling process requires (1) the development of data on LLMW amounts, composition, and shipping routings that are used as inputs to the computational model; (2) determination of the waste-processing module operational parameters used as multiplication factors within the computational model; and (3) operation of the model.

The ANL WASTE_MGMT computational model is run on an IBM-compatible personal computer using the Microsoft FOXPRO® relational database system (4). The model is run using information from three types of data files: a waste volume and contaminant characterization file, a waste-processing module characterization file, and a waste-shipping configuration file.

The computational model tracks the flow of contaminants through each treatment module through the entire treatment train for each waste treatment category. The model utilizes the volume partitioning for each treatment module and the densities of both primary and secondary waste-processing streams to establish a consistent mass balance flow through the entire treatment train for a given waste. The model conserves the waste input mass; added solidification materials (i.e., grout cement) are counted in the final disposal product, but added treatment materials (e.g., process water and incinerator fuel) are not tracked by mass.

In addition to partitioning by volume, the computational model also tracks the partitioning of radionuclide and chemical contaminants through treatment at each waste-processing

module. At each step in the treatment train, a contaminant may partition into the air, liquid, or solid portions of the process. Summing over all the waste streams, the model calculates the mass (and/or concentration) of each contaminant in air, water, and solid (including final disposal product) discharges.

Five major types of LLMW are followed in detail in the model; aqueous liquids, organic liquids, solid process residues, soils, and debris waste. These wastes constitute over 90% of the inventory and projected generation volume of LLMW that will require waste processing as described in the EM PEIS. The current CWMF does not include the processing of the remaining 10% of special category LLMW (e.g., contaminated lead, beryllium, and certain reactive wastes). To facilitate the comparative analyses of waste management costs and risks at each site, the consolidated flowchart was assumed to be site-independent. A particular waste treatment train may contain as few as four or as many as 10 distinct processing modules; moreover, each module may be a part of the treatment sequence for several waste types. The integrated flowchart contains approximately 25 individual treatment modules. The CWMF applies to both Waste Management (WM) inventory and operations LLMW and to LLMW derived from Environmental Restoration (ER) activities. However, material and emissions source terms are modeled separately for the processing of WM- and ER-derived LLMW.

As wastes are transferred between modules in a treatment train, treatment processes may alter the volume and/or mass throughput and the chemical/radiological composition of the waste. The flow of contaminants through the model is driven by how the contaminant chemistry interacts with the specific operational conditions in each processing module. The movement of bulk waste mass is a function of the physical changes in material volume and density through each module. Thus, the flow of contaminants is independent of the bulk mass throughput in the flowchart. Changes in bulk mass between modules are computed by multiplying the processing-induced changes in waste volume by the expected changes in waste density. Changes in the contaminant mass between modules are calculated by multiplying contaminant-specific partitioning coefficients by the input mass of each contaminant. The partitioning of contaminants is dependent on the solubility and volatility of the specific chemical/radiological species, the waste stream matrix, and the operating conditions (e.g., temperature) of each module (technology).

LLMW INPUT DATA

Input data to the computational model consist of (1) site waste stream volumetric data, (2) radioactive waste handling characteristics, (3) waste-treatment-category-specific chemical profiles, (4) site-specific radionuclide profiles, and (5) the waste-shipping configuration that is dependent on the management alternative considered. This section briefly discusses the derivation and limitations of the model input data.

Approximately 128,000 m^3 WM LLMW is in storage (1994 inventory) at 44 DOE sites; another 146,000 m^3 of additional LLMW is expected to be generated through 2014. (An additional 140,000 m^3 of waste from the Hanford and Oak Ridge sites is considered as LLMW but was not modeled for the EM PEIS). Under the semi-restricted treatment case,

ER-derived LLMW is expected to total approximately 7.3 million m^3 through 2030 (5). Contaminant emissions are modeled separately for WM and ER LLMW. For WM LLMW, processing of the total waste volume (inventory plus generated waste) is annualized into an expected 10-year processing period. For ER wastes, the model calculates waste treatment conducted over 30 years.

LLMWs are organized into one of four radiological handling categories depending on the nature and activity of the radioactivity in the waste: contact-handled (CH) non-alpha, CH alpha, remote-handled (RH) alpha, and RH non-alpha. By far the largest volume of waste (>99%) is CH. In the computational model, CH wastes are tracked separately from RH LLMW. The type of radiation emitted also determines LLMW classification and affects emissions modeling. The majority of the modeled LLMW (approximately 75%) is non-alpha waste (less than 10 nCi/g transuranic radionuclide alpha activity). The remaining 25% of LLMW is alpha LLMW (between 10-100 nCi/g transuranic alpha activity) (over 99% of ER LLMW is projected to be CH non-alpha). Because of regulatory concerns about comingling of waste types, contaminant flow and emissions modeling are performed separately for alpha and non-alpha wastes.

For the purpose of the model, the majority of the 2,000 individual MWIR-2 waste streams were grouped into 23 general waste treatment categories (each category is a subset of one of the five major waste types described in the previous section) (6). The 23 waste treatment categories were grouped according to similar chemical/physical treatment characteristics, which allows for common waste processing. Some of the more significant LLMW treatment categories include aqueous waste waters (acidic, basic, neutral, or cyanide), halogenated organic liquids, non-halogenated organic liquids, inorganic particulates and sludges, salt wastes, halogenated organic particulates/sludge, non-halogenated organic sludge, contaminated soils with debris, soils without debris, inorganic non-metal debris, metal debris, combustible debris, heterogeneous debris, and various lab pack wastes. Each waste treatment category follows a distinct treatment train.

Estimation of emission source terms for LLMW requires quantification of the composition and concentration of hazardous chemicals, radionuclides, and certain physical parameters (e.g., ash content) in each waste stream at a site. However, because the current empirical information is insufficient to develop complete chemical/radiological profiles for each waste stream/site combination, the model uses data that assume (1) site-independent chemical profiles based on the generic waste treatment category and (2) site-based radiological profiles that are independent of the particular waste treatment category. The development of LLMW chemical/radiological profiles is detailed in other documents (6, 7, 8).

Chemical profiles for the 23 treatment categories of WM-derived CH non-alpha LLMW were developed from compilation of the chemical concentration data presented in MWIR-2 and an engineering assessment of the industrial processes that generated the respective LLMW streams. Hazardous chemical profiles for ER LLMW were primarily derived from compilation of chemical data from the secondary waste streams expected to be sent to WM for treatment as outlined in the Automated Remedial Assessment Methodology (ARAM) database provided by Pacific Northwest Laboratory (PNL) (5, 9).

Information about historical DOE site operations, industrial processes, and waste generation, suggests that the waste streams of a given treatment category classification at sites across the DOE complex are similar. Thus, the model input data assume that chemical profiles for both WM and ER-derived LLMW are waste-treatment-category-specific and are independent of the site that generated the waste. To provide a consistent assessment of chemicals that is comparable across waste treatment categories, the over 100 individual chemical species identified in DOE LLMW were condensed into 16 distinct chemicals and/or grouped into classes of chemicals that are common to most waste streams. These include six toxic metals (silver, barium, cadmium, chromium, lead, and mercury); three inorganic chemicals (arsenic, selenium, and cyanide); and seven classes of organic chemicals that are grouped according to important treatment parameters such as density, solubility, volatility, and degree of chlorination. These classes are (1) acetone, butanone, and methanol; (2) toluene, xylene, and benzene; (3) trichlorethanes; (4) tetrachloroethanes; (5) dichloroethanes; (6) methylene chloride; and (7) chlorofluoro-hydrocarbons.

The input ash content of a waste stream is related to the amount of particulate emissions from the incineration treatment module. The input ash content is an important component of the LLMW compositional profile because most air releases of hazardous heavy metals and non-soluble radionuclides tend to follow the flow of particulate emissions through the model.

Radiological profiles for LLMW were estimated from the information about the concentrations and activities (Ci) of various radionuclides in LLW compiled in the Integrated Data Base (IDB) (10). The data on LLW radiological profiles were applied to LLMW because it was assumed that the particular radionuclide profiles of LLW and LLMW at a site are dependent on the site operation(s) that generated the radionuclides. The IDB outlines five radiological source profiles that represent the groupings of radionuclides according to the source and time of generation: (1) fission products, (2) induced activity, (3) uranium/thorium, (4) transuranics, and (5) tritium. Each of the DOE sites has a distinct proportion of these five radiological source profiles that is dependent on the site's operational history. A single mission site would be expected to have a radionuclide profile derived primarily from one radiological source category (e.g., uranium-235 concentration facilities like Portsmouth or Paduch have radiological profiles derived almost 100% from the uranium/thorium source profile). Multipurpose sites like Oak Ridge with uranium-235 concentration facilities, reactor operations, and specific isotope production would have an estimated profile that is a proportional combination of the five source profiles (e.g., for Oak Ridge, 30% fission products, 1% uranium/thorium, 1% tritium, and 68% induced activity).

The relative intensities (activity per unit volume in Ci/m^3) of LLMW radionuclides were adjusted from the baseline LLW profiles by "aging" the radionuclides to account for the greater age of LLMW at the assumed time of treatment relative to LLW data in the IDB. The age of LLMW radionuclides is site-specific and depends on the site operational history and whether the LLMW is in current inventory or has yet to be generated. The model uses LLMW radiological profiles aged to the median proposed treatment year of 2008.

The ER LLMW radionuclide profiles correspond to those used for WM LLMW with two adjustments: (1) the mean time for generation of radionuclides in all ER wastes was

assumed to be 1965, which would produce a different adjustment to aging for the ER wastes compared with most WM LLMW at each site, and (2) the intensity of ER LLMW was assumed to be 10% of the intensity for the equivalent WM LLMW at the site. Both the increased time for radioactive decay and dilution of ER wastes by nonradioactive external materials, such as soil and old packaging materials, account for the lower intensity.

Input volumes of LLMW were also adjusted according to one of seven waste-shipping configurations described in the EM PEIS. Each shipping scenario represents a treatment alternative that delineates the sites for LLMW processing. The alternatives range from each site treating its own waste to requiring shipment of all LLMW to the Hanford site for processing. The input information on LLMW volumes, chemical/radiological profiles, and radiation treatment category was adjusted according to the shipping configuration prior to input to the computational model.

FLOWCHART MODEL

The CWMF covers five basic processes: front-end waste handling, pretreatment, primary treatment, secondary treatment, and tertiary (final disposal form) processing (Figure 1).

PLACE FIG. 1 HERE

The waste input is represented by the five waste types (and their 23 subset waste treatment categories). Each of the processing steps consists of one or more modules. Each module may contain one or more specific process technologies. A particular series of modules represents the treatment train for a specific waste treatment category. The arrows in Figure 1 represent the contaminant/waste mass flowpaths through the CWMF. These generic flowpaths apply across sites and for each of the 23 waste treatment categories. The bulk mass partitioning factors along the flowpaths were determined separately for each waste treatment category. The contaminant partitioning coefficients along the flowpaths were determined by how module conditions affect each contaminant and are not dependent on the waste treatment category.

As currently proposed, waste-processing options rely on flame treatment (incineration) for most combustible wastes and grouting as the preferred final waste disposal form. Except for a few existing facilities (e.g., the Liquid Effluent Treatment and Disposal Facility at the Idaho National Engineering Laboratory [INEL]), the processing facilities outlined in the flowchart have yet to be designed. Specific facility operational parameters will be designed on a site-by-site basis. For the computational model, common module operational parameters are used for each generic module, irrespective of site. The flowchart and model are flexible enough to allow for modification of individual waste-processing modules as new waste data, technology information, or DOE waste management guidance are introduced.

The assumed operational conditions of each waste-processing module used in the CWMF are based on empirical data and engineering assessments of similar existing technologies. Proportional waste volume fractions (volume splits) for each module were provided by the MWTP. Waste densities were estimated from data in MWIR-2, along with engineering judgement based on comparison of LLMWs with wastes generated in similar industrial

processes. The chemical/physical partitioning factors are specific for each of the 16 chemical species and depend on the technology used in the module, the waste stream matrix, species solubility/volatility, destruction efficiency, and the residual media (i.e., air emissions, water releases, or incorporation into solid product). For example, the air residual of a particular species (e.g., tri-chlorinated organics) is different in the incinerator module (9.90 E-08 of input is released) than in the wet oxidation module (1.0 E-06 of input). In this case, the factors differ as a result of the effect of the higher incinerator operational temperatures on species volatility and destruction (i.e., the incinerator destroys 99.99% of the tri-chlorinated organics compared to a 99.9% destruction for wet oxidation). A list of the module operational parameters is given in Wilkins et al. (6).

The initial waste mass inputs were determined by multiplying the LLMW volume data with the empirical waste stream gross densities derived from wastes at INEL. The INEL data were used because they represent the most complete information currently available on LLMW densities.

ASSUMPTIONS/UNCERTAINTIES

As described in this paper, two primary factors contribute to uncertainty in the LLMW processing model: (1) the existing DOE data on LLMW are incomplete, and (2) the operational requirements for the CWMF remain more conceptual than specific. To estimate emissions and waste flow source terms, several assumptions were applied to the modeling process. The primary purpose of the assumptions is to provide consistent baseline data that, when run through the computational model, furnish comparative source term results across the DOE sites. The more important assumptions and some of the uncertainty introduced by each are reviewed in this section.

The input data assume different origins for the chemical and radiological contaminant profiles for the LLMW waste streams; chemical profiles are waste-treatment-category-specific, while radionuclide profiles are site-specific. In reality, each of the 2,000 LLMW streams may vary in chemical composition by site and in radiological composition by waste treatment category compared to the generic profiles; however, the current incomplete data do not allow for quantification of these potential differences. In the future, more refined LLMW input data may alter modeled emission output at each site.

The condensing of over 100 chemical contaminants into 16 distinct chemical species and/or chemical groups may oversimplify the differences inherent in how each chemical partitions in a given waste-processing module. For a waste that may contain an unusual species of a given chemical, the model may not provide accurate output data. For example, barium, one of the six toxic metals, is assumed to be water insoluble throughout the model; however, if a particular waste actually contains a soluble form of the chemical (e.g., barium chloride), the partitioning between air and water fractions could be different than that modeled. The generalizing of related compounds into chemical groups can also introduce uncertainty. The model uses volatility and solubility coefficients that represent averaged values for a group of related compounds (e.g., four-chlorine halogenated organics). If a waste contains a preponderance of one particular species in the group, and if the volatility/solubility values of

that species differ from the assumed group averages, the model may give an inaccurate emissions output. With more complete data on LLMW, the model could be refined to account for such differences.

The assumption that waste input density adjustments for all sites are based on data from INEL may introduce uncertainty if the waste composition and, thus, densities of the equivalent LLMW from other sites vary from the INEL data. Because this density adjustment is applied at the front end of the computational model to determine waste input mass, inaccurate density values would affect the mass flow throughout the model. At present, not enough data are available from other sites to make a consistent refinement of the input density adjustment.

The model only tracks the mass balance of input contaminants; emissions from the addition of treatment chemicals are only minimally quantified. For example, the amount of fuel (e.g., fuel oil) needed in the incinerator module is dependent on the heat content of the input waste. The amount of added fuel will affect the emission source terms of such gases as sulfur dioxide (SO_2), nitrogen oxide (NO_x), and carbon monoxide (CO).

Tracking the flow of contaminants independent of the bulk waste mass flow works best when the contaminant mass is a small percentage of the waste mass. For the majority of non-special category LLMW, the contaminant mass constitutes less than 1% of the bulk mass (for inorganic contaminants the value is usually less than 0.01%; certain organic waste streams have organic contaminants concentrations that exceed 1%). A high (greater than 10%) contaminant concentration (especially of an inorganic contaminant that is not destroyed in the treatment process) in a waste stream would result in an inaccurate mass balance through those modules where the chemical partitioning factors differ from the bulk mass partitioning. This would not significantly affect the modeling of the contaminant emission source terms; however, the sum of the contaminant emission mass and the bulk mass output would appear to be greater than the waste input mass. Currently, wastes with known high concentrations of inorganic contaminants (e.g., lead bricks and beryllium wastes) are not run through the model. High concentrations of organic contaminants are less likely to upset the model mass balance because these contaminants are largely destroyed in the treatment process.

The LLMW radiological profiles were assumed to be similar to the profiles for corresponding LLW at each site (adjusted for the age of the waste), as adapted from the IDB (10). This assumption is probably reasonable for the radionuclide profile of LLMW. However, the assumption that the initial intensities (in Ci) of the two waste types are the same may be less accurate. If the initial radiological intensities differed between LLW and LLMW, the level of activity for each radionuclide projected for the model's air emissions, water releases, and residual solids would differ from the current result.

CONCLUSION

The modeling of contaminant and waste material flow and the estimation of emission source terms are important in the effort to understand the risks and costs associated with the proposed alternatives to manage DOE LLMW. The modeling of waste and contaminant

throughput by use of the WASTE_MGMT computational model is one possible method of estimating emission source terms. Clearly, current model results are uncertain because of the assumptions applied to fill in gaps in the existing LLMW data and the conceptual (as opposed to designed) nature of the proposed waste-processing flowchart. Since the proposed waste-processing operations will not be fully realized for several years, changes in management alternatives and regulatory requirements can also affect the validity of the currently modeled data. The modeling process as outlined here is dynamic and can be adapted to future changes in LLMW data, waste-processing module operational criteria, and management alternatives.

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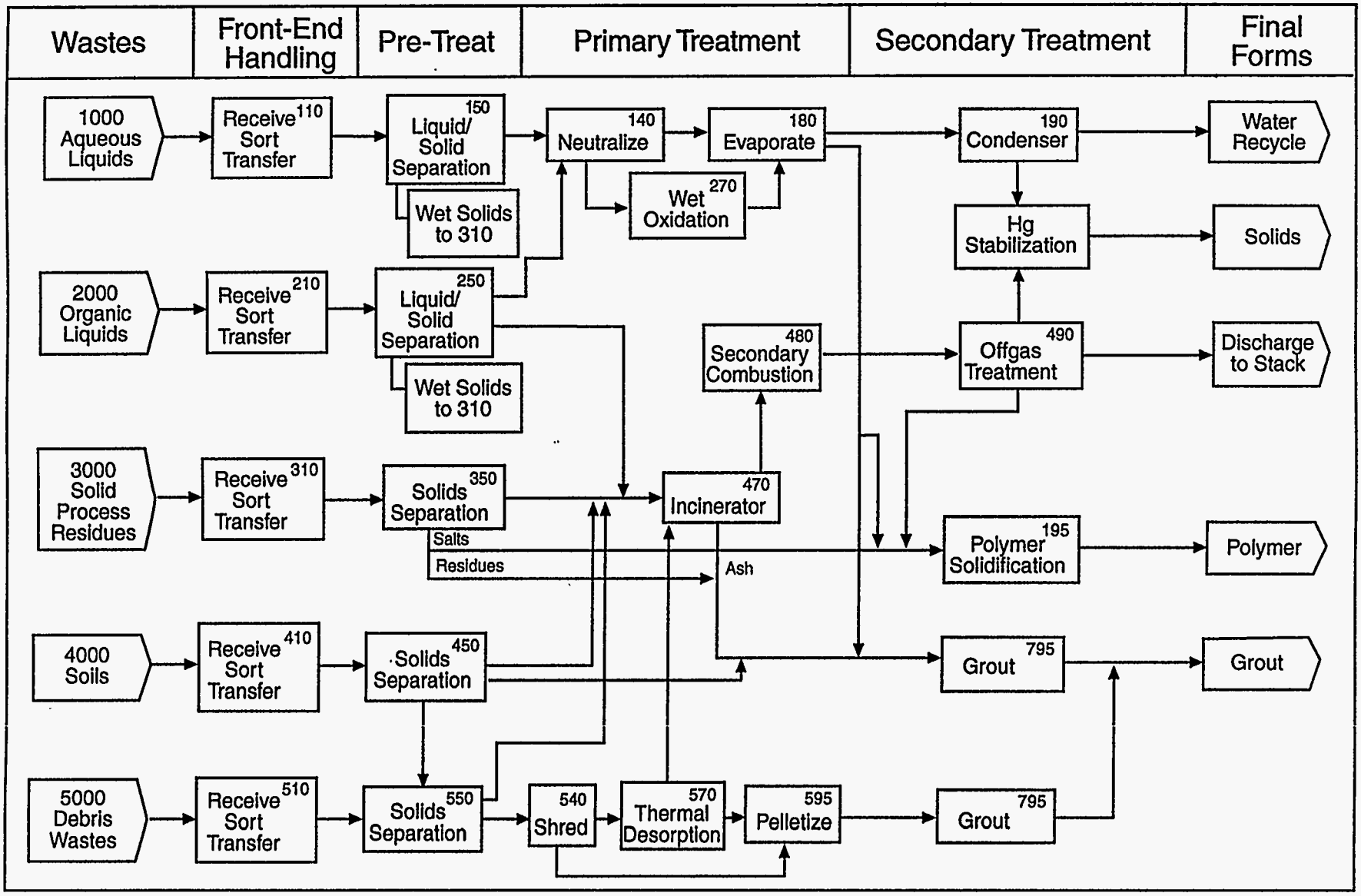
Fig. 1. Consolidated Waste Management Flowchart


Wastes
Front-End Handling
Pre-Treat
Primary Treatment
Secondary Treatment
Final Forms
1000 Aqueous Liquids
Receive 110 Sort Transfer
Liquid/ Solid Separation 150
Wet Solids to 310
Neutralize 140
Wet Oxidation 270
Evaporate 180
Condenser 190
Water Recycle
Hg Stabilization
Solids
2000 Organic Liquids
Receive 210 Sort Transfer
Liquid/ Solid Separation 250
Wet Solids to 310
Secondary Combustion 480
Offgas Treatment 490
Discharge to Stack
3000 Solid Process Residues
Receive 310 Sort Transfer
Solids Separation 350
Salts
Residues
Incinerator 470
Ash
Polymer Solidification 195
Polymer
4000 Soils
Receive 410 Sort Transfer
Solids Separation 450
Grout 795
Grout
5000 Debris Wastes
Receive 510 Sort Transfer
Solids Separation 550
Shred 540
Thermal Desorption 570
Pelletize 595
Grout 795