

MODELING OF LOW-LEVEL-WASTE DISPOSAL FOR ENVIRONMENTAL IMPACT ANALYSIS

Charley Yu
Energy and Environmental Systems Division
Argonne National Laboratory
Argonne, IL 60439

ABSTRACT

A computer model was developed to analyze environmental impacts resulting from disposal of both radioactive and chemical wastes. The model simulates a waste-disposal system and includes infiltration through disposal cell caps, source leach rates, and solute transport in geologic media. The solute transport model used is a modified version of an analytical code originally developed at Oak Ridge National Laboratory (ORNL). The solute transport code was modified based on the Group-Transfer Concentration (GTC) concept developed by the author. The original ORNL model is applicable only to homogeneous media under saturated conditions, but the modified model extends applicability to nonhomogeneous media (layered soils and fractures) and partially saturated conditions. The modified model has broader applicability, e.g., it can be applied to both humid and arid waste-disposal sites, and it is also more practical for environmental impact analysis.

INTRODUCTION

Modeling of low-level-waste disposal became mandatory following issuance in January 1983 of the U.S. Nuclear Regulatory Commission regulations on "Licensing Requirements for Land Disposal of Radioactive Waste" (10 CFR 61). In addition, computer models are usually used to support environmental impact analyses carried out for compliance documents required according to the National Environmental Policy Act (NEPA) of 1969. A computer model was developed by the author to model a waste-disposal system for radioactive as well as chemical wastes; the model includes infiltration through disposal cell caps, source leach rates, and solute transport in geologic media. The solute transport model used is an analytical model originally developed at Oak Ridge National Laboratory (ORNL). The ORNL model was modified based on the Group-Transfer Concentration (GTC) concept so that it can be used in broader situations under more practical conditions. The modeling of a waste-disposal system and the modifications to the original model are discussed in detail in this paper. The application of the modified model to a humid and an arid site are also presented.

MATHEMATICAL MODEL DESCRIPTION

Governing Equation

The migration of wastes in geologic media can be described by nuclide transport equations. Various forms of transport equations have been published in the literature to predict the movement of contaminants in geologic media under various conditions (1-9). A generalized model that can be used under saturated and partially saturated conditions in porous and fractured media has been developed by the author (10). This model, called the Group-Transfer Concentration (GTC) model, has been successfully used to predict tracer movement in large soil columns (11-12) and in field studies (10-13). The GTC model can be written as:

$$\frac{\partial C_i}{\partial t} = \frac{U_{e_i}}{R_{d_i}} \frac{\partial^2 C_i}{\partial Z^2} - \frac{V_i}{R_{d_i}} \frac{\partial C_i}{\partial Z} - \lambda C_i - \frac{K_{e_i}}{R_{d_i}} C_i \quad (1)$$

$$+ \frac{1}{\theta_i R_{d_i}} \sum_{j=1}^n (\alpha_{e_{ij}} C_j - \alpha_{e_{ji}} C_i) + \frac{\dot{M}_i}{R_{d_i}}$$

where for group i:

- D_{e_i} = effective dispersion coefficient ($m^2 \cdot s^{-1}$),
- R_{d_i} = retardation factor = $1 + \frac{f_i \rho_i}{\theta_i} K_{d_i}$, (2)
- K_{e_i} = effective degradation rate (s^{-1}),
- $\alpha_{e_{ij}}, \alpha_{e_{ji}}$ = effective group mass transfer coefficient ($m^3 \cdot m^{-3} \cdot s^{-1}$),
- θ_i = volumetric water content in group i ($m^3 \cdot m^{-3}$),
- C_i, C_j = dissolved concentration in group i and j, respectively ($kg \cdot m^{-3}$),
- f_i = fraction of bulk volume associated with group i,
- ρ_i = bulk density of the media associated with group i ($kg \cdot m^{-3}$),
- K_{d_i} = distribution coefficient for group i ($m^3 \cdot kg^{-1}$),
- v_i = average pore water velocity in group i ($m \cdot s^{-1}$),
- λ = radioactive decay constant (s^{-1}),
- \dot{M}_i = source input in group i ($kg \cdot m^{-3} \cdot s^{-1}$),

t = time (s), and

Z = distance along the flow path (m).

Equation (1) was solved numerically using the finite-differences method. A numerical dispersion problem was experienced and was solved by analyzing the truncation error of the Taylor series expansion of the advection term (10, 14).

The concept of the GTC model was used to modify an analytical model developed at Oak Ridge National Laboratory (15). The analytical model, called AT123D, solves a transport equation similar to Eq. (1), but with the group number i set equal to unity and the transfer coefficients $\alpha_{e_{ij}}$ and $\alpha_{e_{ji}}$ set equal to zero. The transport equation is solved analytically in AT123D using Green's function method for various boundary conditions (15). The AT123D model is applicable only to homogeneous media under saturated conditions. For purposes of environmental impact analysis, AT123D was modified based on the GTC concept so that it can have broader applicability to more realistic geohydrologic conditions.

The modifications of AT123D include (a) incorporating a transient waste-leaching option, (b) extending the application of the model to saturated-unsaturated media, and (c) extending the application of the model to macropore/fracture conditions. The modified version computes in double precision whereas the original AT123D was developed in single precision. Detailed descriptions of the modifications follow.

Transient Waste Leaching

The waste release rate (source strength) is calculated using a first-order leaching rate (exponential leaching) model (16). The source strength, $\dot{Q}(t)$, can be written as

$$\dot{Q}(t) = L_d \cdot \rho_b \cdot V \cdot C_d(t) \quad (3)$$

where L_d is the solute leach rate, ρ_b is the waste density, V is the waste volume, and $C_d(t)$ is the solute concentration in the wastes available for leaching at any time t. The solute leach rate, L_d , can be calculated (16) as

$$L_d = \frac{I}{\theta_e L_i R_d} \quad (4)$$

where I is the infiltration rate, θ_e is the effective moisture content in the wastes, L_i is the waste leaching thickness, and R_d is the solute retardation factor in the wastes. The effective moisture content, θ_e , in Eq. (4) is the product of effective porosity, n_e , and the moisture saturation ratio, R_s ; R_s is defined as the fraction of the voids in a porous medium that is filled with water. The infiltration rate and retardation factor are discussed later.

The solute concentration at any time t, $C_d(t)$, is obtained by solving the following two differential equations

$$\frac{dC_p(t)}{dt} = -(\lambda_p + L_p) C_p(t) \quad (5)$$

and

$$\frac{dC_d(t)}{dt} = -(\lambda_d + L_d) C_d(t) + \lambda_d C_p(t) \quad (6)$$

where $C_p(t)$ is the parent radionuclide concentration, λ_p is the parent decay constant, L_p is the parent leach rate, and λ_d is the daughter (radionuclide of interest) decay constant. For radionuclides with L_p much less than λ_p , as in the example discussed below, Eq. (5) reduces to

$$\frac{dC_p(t)}{dt} = -\lambda_p C_p(t) \quad (7)$$

Solving Eqs. (6) and (7) with initial source concentrations ($\rho Ci/g$) of S_p and S_d for parent and daughter radionuclides, respectively, one obtains

$$C_d(t) = S_d e^{-(L_d + \lambda_d)t} + \frac{\lambda_d S_p}{L_d + \lambda_d - \lambda_p} [e^{-\lambda_p t} - e^{-(L_d + \lambda_d)t}] \quad (8)$$

If the daughter nuclide source concentration reaches a maximum value, $C_d(t_{max})$, at time t_{max} after waste emplacement, the time to reach the maximum source concentration can be calculated by

$$t_{max} = \frac{1}{\lambda_d + L_d - \lambda_p} \cdot \ln \left[\left(\frac{\lambda_d + L_d}{\lambda_p} \right) \left(1 - \frac{S_d}{S_p} \cdot \frac{\lambda_d + L_d - \lambda_p}{\lambda_d} \right) \right] \quad (9)$$

The maximum source concentration, calculated by substituting the t_{max} calculated from Eq. (9) into Eq. (8), is therefore $C_d(t_{max})$.

Saturated-Unsaturated Media

The original AT123D model can be used only for saturated conditions. The AT123D model was modified so that it can be used for saturated-unsaturated media as illustrated in Fig. 1. The modification used a travel-time method described by Gilbert et al. (17). The waste field was assumed to be a rectangular block with dimensions L_x , L_y , and L_z . The geometry of the contaminated layer was assumed to remain constant during vertical migration in the unsaturated (vadose) zone. Precipitation falling on the waste field would infiltrate through the wastes and would transport contaminants down through the unsaturated (vadose) zone into the saturated (aquifer) zone. After the contamination reached the water table, it would spread out, be diluted in the groundwater, and generate a contaminant plume that would move in the downgradient direction.

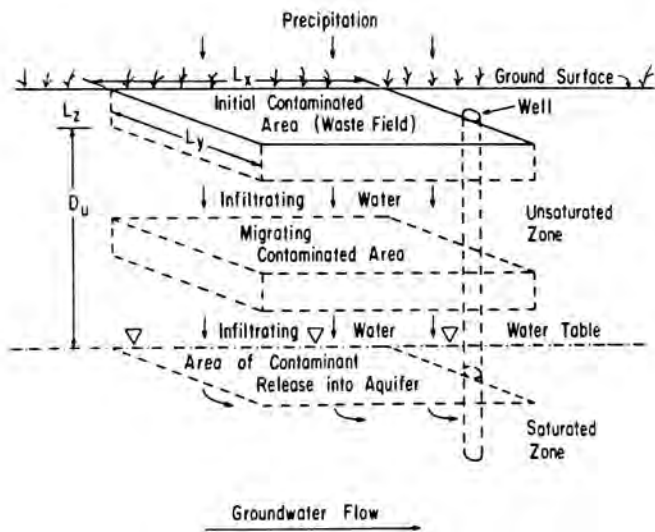


Fig. 1. Schematic Diagram of Model for Contaminant Transport in Saturated-Unsaturated Media. Source: Modified from Ref. 18.

The travel-time method is extended to handle layered soil conditions in the vadose zone. The travel time, i.e., the time for contaminants to be transported through the vadose zone, for an N-layered zone is calculated as

$$T_d = \sum_{k=1}^N L_k \theta_{e_k} R_{d_k} / I \quad (10)$$

where T_d is the travel time, L_k is the k th layer thickness, θ_{e_k} is the k th layer effective moisture content, R_{d_k} is the k th layer retardation factor, and I is the infiltration rate. The retardation factor and infiltration rate are discussed later.

Radioactive decay during transport through the vadose zone is considered in calculating the source term. The concentration of the radionuclide is multiplied by $\exp(-\lambda_d T_d)$, where λ_d is the radioactive decay constant and T_d is the travel time through the vadose zone.

Macropore and Fracture Conditions

The AT123D model was originally developed for a homogeneous medium. In order to broaden the applicability of the model to more realistic situations for a typical waste-disposal system, the model was modified so that it can be used under macropore/fracture conditions. The modification was based on the GTC concept, which resulted in modification of the retardation factor used in the AT123D model. The retardation factor is calculated in AT123D using the equation

$$R_d = 1 + \rho_b K_d / n_e \quad (11)$$

where ρ_b is the bulk density of the medium (kg/m^3), K_d is the distribution coefficient, and n_e is the

effective porosity. Based on the GTC model, bulk density in Eq. (11) should be replaced by effective density, which is defined as the quotient of the solid mass in the effective region divided by the bulk volume. The solid mass in the effective region is defined in the GTC model as the solid mass associated with the group of interest. Because the solid mass in the effective region is always less than or equal to the bulk mass, the retardation factor calculated using Eq. (11) is an overestimate (i.e., nonconservative). An alternative approach is therefore required.

The GTC model defines the retardation factor for an arbitrary group, group i (e.g., the solution channels) (10, 12), as

$$R_{d_i} = 1 + \frac{f_i \rho_i}{\theta_i} K_{d_i} \quad (12)$$

where f_i is the fraction of bulk volume associated with group i , ρ_i is the bulk density of the medium associated with group i , θ_i is the volumetric water content in group i , and K_{d_i} is the distribution coefficient for group i . It can be shown (10, 12) that

$$\sum_{i=1}^n f_i \rho_i = \rho_b \quad (13)$$

and

$$\sum_{i=1}^n \theta_i = \theta_t \quad (14)$$

where n is the total number of groups, ρ_b is the bulk density of the entire medium, and θ_t is the total water content in the medium. Because the application of AT123D to a medium containing macropores or fractures is essentially the same as using the single-group option of the GTC model to solve a multigroup problem (medium containing macropores/fractures) (12), a set of effective parameters should be chosen to reflect this effect and to obtain conservative estimates of contaminant concentrations. For $n = 1$ (single group), $f_i \rho_i = \rho_b$ and $\theta_i = \theta_t$. That is,

$$R_d = 1 + \frac{\rho_b}{\theta_t} K_d = 1 + \frac{\rho_b K_d}{n_t R_s} \quad (15)$$

where n_t and R_s are the total porosity and saturation ratio of the medium, respectively.

When estimating contaminant concentrations in a medium containing macropores or fractures, Eq. (15) is a more conservative way of calculating the retardation factor than Eq. (11). Equation (15) has been incorporated in the modified version of AT123D. The modified AT123D will be referred to as MAT123D in the remaining discussions.

MODEL APPLICATIONS

To illustrate the applications of the MAT123D model to environmental impact analysis, two typical waste-disposal sites were chosen as examples. One, Site A, is located in a relatively humid environment; the other, Site B, is located in a relatively arid environment.

The disposal of low-level wastes and some long-lived radioactive wastes -- such as raffinates, tailings, and contaminated soil materials containing uranium- and thorium-series radionuclides -- often requires greater-confinement disposal (GCD), which is disposal that provides a degree of isolation greater than conventional shallow-land burial (SLB) (19). The need for GCD has resulted not only because of the existence of wastes exceeding regulatory limits for SLB but also because of individual policies of organizations and demands of concerned citizens (19). In the examples discussed below, the waste-disposal system includes a multilayered cap of natural materials to resist the infiltration of water. Examples of the latest conceptual designs for such units are given in various reports (18, 20-22).

The infiltration rates through the cover materials are calculated based on estimated runoff coefficients using the equation (17)

$$I = (1 - C_r) P - E \quad (16)$$

where I is the infiltration rate, C_r is the runoff coefficient, P is the annual precipitation, and E is the evapotranspiration rate. The runoff coefficients can be estimated from the slope and properties of the cover materials. The waste stream considered in the example is composed of three long-lived isotopes, i.e., uranium-238, thorium-230, and radium-226. Because thorium is highly adsorbed by soils with an average distribution coefficient of 60,000 mL/g (17), only uranium-238 and radium-226 are analyzed using the MAT123D model. However, the ingrowth of radium-226 from radioactive decay of thorium-230 is considered in the waste-leaching calculations using Eq. (8).

The output of the MAT123D lists the travel times of water and all contaminants considered. It also lists the contaminant concentrations as functions of time and space. The travel times calculated by MAT123D for Sites A and B are presented in Table I. The calculated water travel times are 1.2 years for the humid Site A and 260 years for the arid Site B. The calculated radionuclide travel times for both sites are greater than 3,900 years for uranium-238 and greater than 13,000 years for radium-226. The radionuclide travel time is greater than the water travel time due to the sorptive properties of soil. If using the retardation factor defined in AT123D, i.e., Eq. (11), the radionuclide travel times for the Site containing fractures (Site A) will be about 10 times greater than that calculated by MAT123D using Eqs. (10) and (15). Therefore, for purposes of environmental impact analysis, MAT123D is more conservative than the original ORNL model.

The time and spatial variations of radium-226 and uranium-238 concentrations in groundwater were calculated by the MAT123D model. The radionuclide concentrations in the hypothetical on-site well at the waste-field boundary and the times to reach maximum concentrations are presented in Table II. All parameters used in MAT123D to generate the results listed in Tables I and II are listed in Table III. The calculated maximum concentrations of uranium-238 and radium-226 are low for both Site A and Site B. The times to reach maximum concentrations for uranium-238 and radium-226 are greater than 3,900 years for both sites. However, for uranium-238 and radium-226, the maximum concentrations at the relatively humid site (Site A) are greater than that

at the relatively arid site (Site B) whereas the times to reach maximum concentrations are much greater at the arid site than at the humid site.

TABLE I
Water and Radionuclide Travel Times

Site	Travel Time (yr)		
	Water ^a	Uranium-238	Radium-226
A (humid)	1.2	3,900	13,000
B (arid)	260	20,000	52,000

^a Calculated using Eqs. (10) and (15), with R_d equal to unity.

TABLE II
Radionuclides in Hypothetical On-site Wells^a

Site	Uranium-238	
	Maximum Concentration (pCi/L)	Time to Reach Maximum Concentration ^b (yr)
A (humid)	0.22	3,900
B (arid)	0.18	20,000

Site	Radium-226	
	Maximum Concentration (pCi/L)	Time to Reach Maximum Concentration ^b (yr)
A (humid)	0.0004	22,000
B (arid)	0.00005	61,000

^a Wells are assumed to be located next to the waste-disposal area.

^b Includes the travel times for nuclides to move from the bottom of the waste field to the water table (see Table I for radionuclide travel times).

CONCLUSION

The modified model, MAT123D, extends applicability of the original model to more realistic conditions for disposal of low-level waste. The MAT123D model can be successfully applied to a disposal system situated in saturated-unsaturated media under macropore/fracture conditions. It can be used to compare environmental impacts for alternative sites and to compare the environmental impacts of various engineering designs at the same site. The modifications to the model not only broaden the applicability of the original model but also make the model more practical for environmental impact analysis.

TABLE III

Parameters Used in the MAT123D Model

Parameter	Symbol	Unit ^a	Humid Site A	Arid Site B
Annual precipitation	P	m/yr	0.94	0.16
Evapotranspiration rate	E	m/yr	0.84	0.10
Runoff coefficient	C _r	-	0.053	0.31
Infiltration rate	I	m/yr	0.05	0.01
Total porosity:	n _t	-		
Wastes			0.23	0.23
Unsaturated zone				
Layer 1			0.42	0.40
Layer 2			0.42	_b
Saturated zone			0.30	0.40
Effective porosity:	n _e	-		
Wastes			0.23	0.23
Unsaturated zone				
Layer 1			0.05	0.2
Layer 2			0.005	_b
Saturated zone			0.0015	0.2
Dry bulk density:	ρ _b	kg/m ³		
Wastes			1,700	2,100
Unsaturated zone				
Layer 1			1,800	1,600
Layer 2			1,800	_b
Saturated zone			2,400	1,600
Hydraulic gradient	i	-	0.0095	0.002
Hydraulic conductivity	K	m/yr	1.2 × 10 ²	6.0 × 10 ⁴
Longitudinal dispersivity	α _L	m	10	3.0
Transverse dispersivity	α _T	m	1.0	0.3
Vertical dispersivity	α _v	m	1.0	0.3
Unsaturated zone depth ^b :	D _u	m		
Layer 1			2	65
Layer 2			4	0
Saturation ratio:	R _s	-		
Wastes			1.0	1.0
Unsaturated zone				
Layer 1			0.5	0.2
Layer 2			0.5	_b
Saturated zone			1.0	1.0
Source concentration:	S	pCi/g		
Uranium-238			120	220
Thorium-230			1,500	2,800
Radium-226			56	110
Distribution coefficient:	K _d	mL/g		
Uranium--				
Wastes			370	370
Unsaturated zone				
Layer 1			370	3.7
Layer 2			370	_b
Saturated zone			0	3.7
Radium--				
Wastes			1,200	1,200
Unsaturated zone				
Layer 1			1,200	10
Layer 2			1,200	_b
Saturated zone			100	10
Waste field dimensions:				
Length	L _x	m	240	240
Width	L _y	m	240	240
Depth	L _z	m	5.7	2.4

^a A hyphen means that the parameter is dimensionless.

^b The unsaturated zone has two layers for Site A and one layer for Site B.

REFERENCES

1. K.H. COATS and B.D. SMITH, "Dead-End Pore Volume and Dispersion in Porous Media," *Soc. Pet. Eng. J.*, 4, 73 (1964).
2. J. SKOPP and A.W. WARRICK, "A Two-Phase Model for the Miscible Displacement of Reactive Solutes through Soils," *Soil Sci. Soc. Am. Proc.*, 38, 545 (1974).
3. M.TH. VAN GUNECHTEN and P.J. WIERENGA, "Mass Transfer Studies in Sorbing Porous Media. I. Analytical Solutions," *Soil Sci. Soc. Am. J.*, 40, 473 (1976).
4. R.A. FREEZE and J.A. CHERRY, *Groundwater*, Prentice-Hall, Inc., Englewood Cliffs, New Jersey (1979).
5. J.P. GAUDET, H. JEGAT, G. VANCHAUD, and P.J. WIERENGA, "Solute Transfer, with Exchange between Mobile and Stagnant Water, through Unsaturated Sand," *Soil Sci. Soc. Am. J.*, 41, 665 (1977).
6. P.S.C. RAO, R.E. JESSUP, D.E. ROLSTON, J.M. DAVIDSON, and D.P. KILCREASE, "Experimental and Mathematical Description of Non-absorbed Solute Transfer by Diffusion in Spherical Aggregates," *Soil Sci. Soc. Am. J.*, 44, 684 (1980).
7. G.E. GRISAK and J.F. PICKENS, "Solute Transport through Fractured Media: 1. The Effect of Matrix Diffusion," *Water Resour. Res.*, 16, 719 (1980).
8. D.H. TANG, E.O. FRIND, and E.A. STUDICKY, "Contaminant Transport in Fractured Porous Media: Analytical Solution for a Single Fracture," *Water Resour. Res.*, 17, 555 (1981).
9. C. YU, W.A. JESTER, and A.R. JARRETT, "Simultaneous Determination of Dispersion Coefficients and Retardation Factors for a Low Level Radioactive Waste Burial Site," *Radioact. Waste Manage. Nucl. Fuel Cycle*, 4, 401 (1984).
10. C. YU, "Mathematical Evaluation of Effective Hydrogeologic Parameters and Media Nonhomogeneity from Tracer Breakthrough Curve Data," Ph.D. Thesis, Pennsylvania State University, University Park, Pennsylvania (1984).
11. C. YU, W.A. JESTER, and A.R. JARRETT, "Hydrogeologic Parameter Identification from Nuclear Tracer Breakthrough Curve Data for Waste Disposal in Porous Media," *Radioact. Waste Manage. Nucl. Fuel Cycle*, 7, 63 (1986).
12. C. YU, W.A. JESTER, and A.R. JARRETT, "A General Solute Transport Model and its Applications in Contaminant Migration Analysis," *Proc. National Water Well Assoc. Conf. on Practical Applications of Groundwater Models*, Worthington, Ohio, p. 353 (1985).
13. C. YU, W.A. JESTER, and A.R. JARRETT, "Determination of Hydrogeologic Parameter and Media Nonhomogeneity from Nuclear Tracer Breakthrough Curve Data," *Proc. 5th Int. Conf. on Nuclear Methods in Environmental and Energy Research*, CONF-840408, p. 277 (1984).
14. D.W. PEACEMAN, *Fundamentals of Numerical Reservoir Simulation*, Elsevier Scientific Publishing Company, New York (1977, Reprinted 1983).
15. G.T. YEH, "AT123D: Analytical Transient One-, Two-, and Three-Dimensional Simulation of Waste Transport in the Aquifer System," ORNL-5602, Oak Ridge National Laboratory, Oak Ridge, Tennessee (1981).
16. C.B. BAES, III, and R.D. SHARP, "A Proposal for Estimation of Soil Leaching and Leaching Constants for Use in Assessment Models," *J. Environ. Qual.* 12, 17 (1983).
17. T.L. GILBERT, et al., "Pathways Analysis and Radiation Dose Estimates for Radioactive Residues at Formerly Utilized MED/AEC Sites," ORO-832 (Rev.), Prepared for U.S. Department of Energy by Argonne National Laboratory, Argonne, Illinois (1983, Rev. 1984).
18. U.S. DEPARTMENT OF ENERGY, "Final Environmental Impact Statement, Long-Term Management of the Existing Radioactive Wastes and Residues at the Niagara Falls Storage Site," DOE/EIS-0109F, Washington, D.C. (1986).
19. L.E. TREVORROW, T.L. GILBERT, C. LUNER, P.A. MERRY-LIBBY, N.K. MESHKOV, and C. YU, "Greater-Confinement Disposal of Radioactive Wastes," *Waste Management '85, Proc. Symposium*, Tucson, Arizona, March 24-28, 1985, p. 127 (1985).
20. U.S. DEPARTMENT OF ENERGY, "Engineering Evaluation of Alternatives for the Disposition of the Weldon Spring Raffinate Pits Site," DOE/OR/20722-5, Prepared by Bechtel National, Inc., Oak Ridge, Tennessee (1984).
21. U.S. NUCLEAR REGULATORY COMMISSION, "Final Environmental Impact Statement Related to the Decommissioning of the Rare Earths Facility, West Chicago, Illinois, Docket No. 40-2061," NUREG-0904, Office of Nuclear Material Safety and Safeguards (1983).
22. U.S. DEPARTMENT OF ENERGY, "Remedial Action Plan for Stabilization of the Inactive Uranium Mill Tailings Site at Canonsburg, Pennsylvania," UMTRA-DOE/AL-140, Albuquerque Operations Office, Albuquerque, New Mexico (1983).