

COMPUTER SIMULATION OF THE CHEMICAL TREATMENT OF A CONTAMINATED GROUNDWATER USING GEOCHEMICAL MODELING TECHNIQUES

Robert W. Sobocinski and Jonathan Myers
IT Corporation
Albuquerque, New Mexico

ABSTRACT

The treatment of a radionuclide- and heavy-metal-contaminated groundwater by the addition of lime is simulated using the EQ3NR/EQ6 geochemical code. The simulation demonstrates the use of geochemical modeling techniques in evaluating water treatment alternatives and optimizing laboratory bench-scale tests. Results indicate that lime treatment is effective in reducing americium, neptunium, uranium, chromium, lead, and silver concentrations but not cesium, strontium, arsenic, or selenium concentrations. Geochemical modeling determined the optimal pH range for the removal of specific contaminants. Modeling also estimated the mass of lime per liter of contaminated groundwater required for treatment and the mass and composition of solid waste material formed during the precipitation process. This study shows that geochemical modeling techniques can effectively be used to evaluate, design, and optimize chemical processes considered for the removal of radionuclides and heavy metals from contaminated groundwater or process waste streams.

INTRODUCTION

Radionuclides and heavy metals are groundwater contaminants at both active and inactive government-owned and private facilities. The effectiveness of proposed treatment techniques is commonly estimated in groundwater remediation/feasibility studies. One such technique, pH adjustment, followed by gravity settling or filtration, can effectively decrease heavy metal concentrations in groundwater (1). Process aqueous wastes are similarly treated to create acceptable immobilized waste forms and to minimize contaminants in wastewater. Neutralization/filtration and caustic coprecipitation are utilized to remove actinide contaminants from aqueous process waste at the U.S. Department of Energy (DOE) Rocky Flats Plant (2). Typically, laboratory bench-scale tests are performed to evaluate the design and optimize the efficiency of the full-scale treatment operation. The treatment process selected for full-scale use is usually the most cost-effective method that will decrease contaminant concentrations in the effluent to acceptable levels.

Computer simulation techniques provide a cost-effective method of screening treatment alternatives because the techniques can simulate the processes used to remove radionuclides and heavy metals from process waste streams or contaminated groundwater. Processes such as pH adjustment, oxidation/reduction, and ion exchange can easily be simulated using speciation and reaction-path modeling techniques that allow for the evaluation and optimization of the treatment method. These simulations can reduce or focus bench-scale testing, and thereby, reduce the high costs associated with these tests.

This study illustrates the use of geochemical modeling techniques in evaluating the effectiveness of the pH adjustment/precipitation process on a hypothetical groundwater contaminated with radionuclides and heavy metals. The incremental addition of lime (CaO), subsequent pH increase, and precipitation of radionuclide- and heavy-metal-bearing solids are modeled using the EQ3NR/EQ6 geochemical code (3,4).

GEOCHEMICAL MODELING

Most, if not all, computer programs designed to model mineral/water interactions are based on the principal of equilibrium thermodynamics. These models require a data base

containing the thermodynamic properties of elements, aqueous species, and minerals. Several programs have been developed to model chemical solubility, speciation, and reaction paths in aqueous solutions. These programs include REDEQL (5,6), MINEQL (7), MINTEQA2 (8), SOLVEQ (9,10), PHREEQE (11), and EQ3NR/EQ6 (3,4). Contaminant solubilities, i.e., maximum concentrations that can exist in solution, and contaminant speciation can be obtained from geochemical modeling, and the progress of reactions that change solution chemistry can be predicted by reaction-path modeling.

Geochemical codes require a complete description of the solution composition to model solubility and speciation. The description should include all major cations and anions, minor cations and anions of interest, pH, total dissolved solids, specific gravity, temperature, and redox. The redox can be expressed as either Eh, pe, oxygen fugacity, or concentrations of specific redox couples. In addition to the above data requirements, the reaction of a solution with solids requires information on the mineralogical composition of the solids and relative proportions of the minerals.

The EQ3NR/EQ6 geochemical codes are solubility/speciation/reaction-path computer programs developed by Lawrence Livermore National Laboratory (3,4). The codes are generally considered to be industry standards and have widespread use in mineral exploration (12,13) and hazardous waste disposal. The codes were used to predict the solubility of radionuclides by Kerrisk (14) and Ogard and Kerrisk (15). EQ3NR/EQ6 was also used to model the mobility of technetium and ruthenium by Coles and Ramspot (16) and Isherwood (17).

EQ3NR/EQ6 has been validated using standard geochemistry problems such as the speciation of sea water (18), basalt/seawater interactions (19), and numerous comparisons with experimentally-determined mineral solubilities (20). Benchmark comparisons with the results of similar codes have been performed by INTERA (21), Nordstrom (18), Kincaid and Morey (22), and Kerrisk (23).

MODELING PROCEDURE

Table I shows a hypothetical groundwater composition with elevated radionuclide and heavy-metal concentrations.

Most radionuclide concentrations were solubility-limited. The uranium concentration was arbitrarily set at ten times the proposed health-based limit, and the cesium and strontium concentrations were both arbitrarily set equal to 1 milligram per liter (mg L^{-1}). Each heavy-metal concentration was either solubility-limited or arbitrarily set equal to some multiple of the drinking water maximum contaminant level (MCL), as specified by the U.S. Environmental Protection Agency (EPA) Safe Drinking Water Act (24). Except for the elevated levels of radionuclides and heavy metals, the groundwater composition in Table I is representative of fresh, near-surface groundwater. Calcium and carbonate species are the dominant dissolved constituents. The initial pH is slightly lower than would be expected, i.e., 5.2 versus 7 ± 1 . The lower pH was required to maintain elevated concentrations of heavy metals and radionuclides in solution. The initial redox potential was set at 579 mV. This value is typical of the relatively oxidizing conditions that exist in shallow aquifers with appreciable hydraulic connection with the surface, i.e., aquifers that are susceptible to contamination. Temperature was set at 11° Celsius (C).

Lime is added to the solution at a fixed rate, and the pH and Eh of the solution are allowed to vary during the simulation. However, the temperature is fixed. Also, the simulation is closed, i.e., no components (such as oxygen) are allowed into or out of the system. As the lime treatment progresses, the speciation and solubility of all dissolved elements are monitored. If the solubility of a dissolved element is exceeded, it is allowed to precipitate, and the resulting solution composition and speciation are recalculated. Solids that precipitate during the simulation are not removed and are available to redissolve as the simulation proceeds.

RESULTS

Simulation results include a tabulation of radionuclide and heavy-metal concentrations and speciation, pH, and Eh as a function of the cumulative mass of lime added to solution. Also included is the cumulative mass of each mineral that has precipitated from solution in response to the addition of lime. Table II shows the results of the simulated groundwater treatment. The addition of lime was quite effective in lowering the concentrations of several radionuclides and metals. Concentrations of americium, neptunium, plutonium, and uranium were lower in the effluent than prior to treatment; cesium and strontium concentrations were not affected by the lime treatment (Table II). In addition, concentrations of chromium, lead, mercury, and silver were also lowered by the lime treatment. However, arsenic, and selenium concentrations were not affected by the treatment. Table II shows that although concentrations were slightly lower in the effluent, lime treatment did not effectively remove plutonium or mercury from the groundwater.

Figure 1 shows the pH as a function of the moles of lime added to the groundwater. It is apparent from Fig. 1 that the pH stabilized after the addition of approximately 0.022 moles per liter (mol L^{-1}) or 1.23 grams per liter (g L^{-1}) of lime. Steady-state conditions occurred at pH 12.9, when the mass of calcium supplied to the groundwater by the addition of lime was balanced by the precipitation of portlandite [$\text{Ca}(\text{OH})_2$] and calcite (CaCO_3). The simulation shows that further pH adjustment by the addition of lime was not possible.

TABLE I

Groundwater Composition For Lime Treatment Simulation

Parameter	Concentration (mg L^{-1})	
Radionuclides		
americium	40.5	solubility limit
cesium	1.0	
neptunium	2.08×10^{-4}	solubility limit
plutonium	6.51×10^{-12}	solubility limit
strontium	1.0	
uranium	0.33	10 x proposed limit
Metals		
arsenic	2.5	50 x MCL
calcium	90.6	
chromium	2.29×10^{-3}	solubility limit
iron	2.82×10^{-6}	
lead	2.5	50 x MCL
mercury	7.16×10^{-8}	solubility limit
potassium	5.58	
magnesium	24.3	
manganese	0.01	
selenium	1.0	100 x MCL
silver	0.025	solubility limit
sodium	13.6	
Anions		
total carbonate (as HCO_3^-)	274.3	
chloride	185.0	
fluoride	0.48	
nitrate	0.34	
phosphate	0.09	
sulfate	107.4	
pH	5.2	
Eh	579 mV	
Temperature	11.0°C	

Figure 2 shows radionuclide concentrations in solution as a function of the pH of the treated groundwater. Only those radionuclides that were affected by the lime treatment are included in Fig. 2.

Each of the radionuclides responded somewhat differently to the addition of lime. The simulated treatment decreased the americium concentration in groundwater by over four orders of magnitude (Table II). The americium concentration decreased steadily from the beginning of the simulation, to a minimum at pH 9.1 to 9.4 (Fig. 2a). With increasing pH, from the initial value of 5.2 (Table I) to approximately 9.4, the bicarbonate ion (HCO_3^-) became the dominant species of dissolved carbonate. Since the americium-bearing precipitate, $\text{AmO}(\text{HCO}_3)$, contains bicarbonate, americium concentrations were lowest in the pH range where HCO_3^- predominates. Beyond pH 9.4, the carbonate equilibria shifted toward carbonate ion (CO_3^{2-}), allowing the $\text{AmO}(\text{HCO}_3)$ to redissolve and americium concentrations to increase. Americium concentrations did not, however, continue to rise as the simulation proceeded beyond pH 9.4 (Fig. 2a). This was because another americium-bearing solid [$\text{Am}(\text{OH})_3$] began to precipitate and control the americium concentration. The simulated lime treatment also decreased

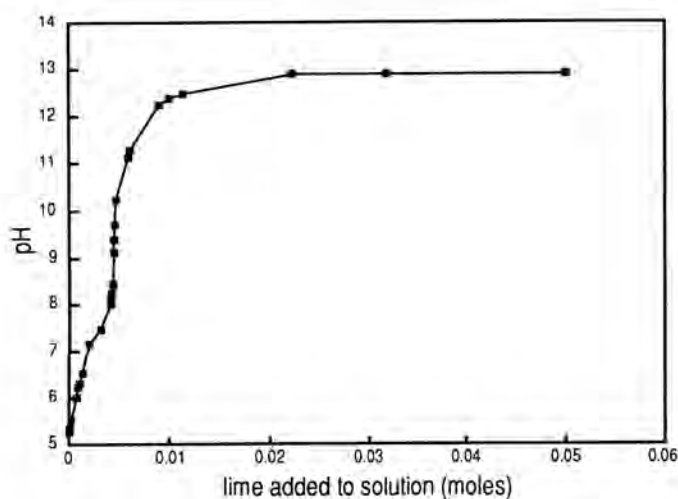


Fig. 1. pH versus moles of lime added to solution.

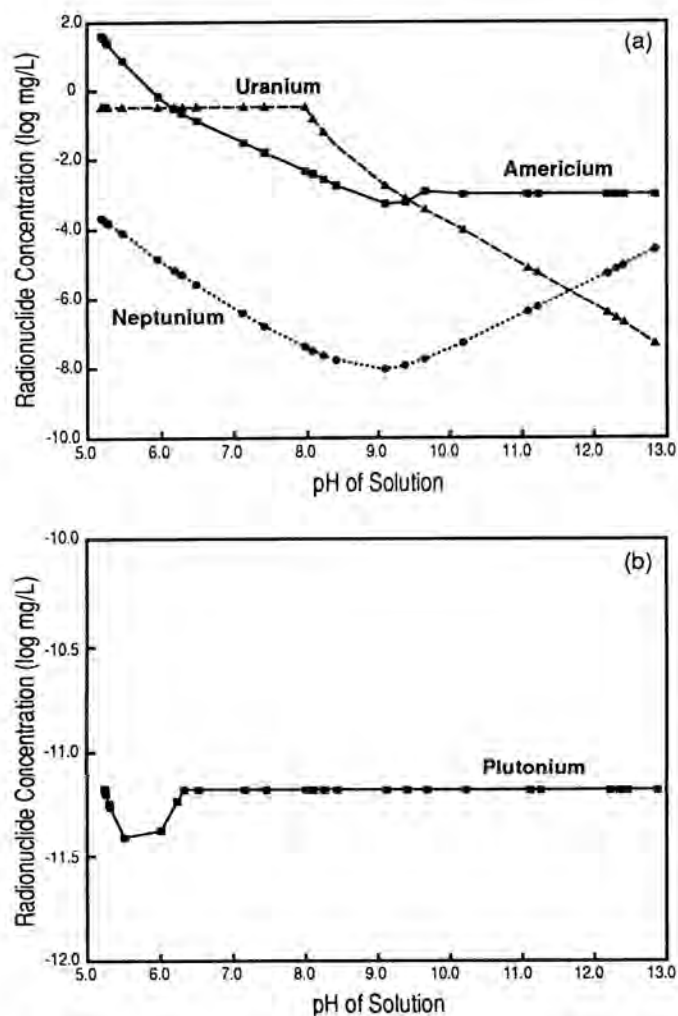


Fig. 2. Radionuclide concentrations versus pH of solution.

the neptunium concentration by over four orders of magnitude (Table II). The concentration of neptunium reached a minimum at pH 9.1 (Fig. 2a). This was due to the multi-valent

nature of neptunium and the decrease in the Eh of the groundwater, from 579 to 92 mV, as the simulation proceeded. The neptunium-bearing solid (NpO_2) contains neptunium in the +4 valence state (Np^{+4}). The maximum concentration of Np^{+4} occurred at pH 9.1, and thus NpO_2 was most stable at this pH. Plutonium concentrations were very low at the start of the simulation (Table I). The plutonium concentration decreased slightly during the simulation, reaching a minimum at pH 5.5 to 6.0 (Fig. 2b). The behavior of plutonium in the treatment simulation was analogous to that of neptunium; plutonium is multi-valent. The maximum concentration of plutonium in the +4 valence state occurred in the pH range of 5.5 to 6.0, and thus, PuO_2 was most stable at this pH range. The simulated lime treatment decreased the concentration of uranium in groundwater by over six orders of magnitude (Table II). Beginning at approximately pH 8.0, the uranium concentration decreased throughout the treatment simulation (Fig. 2a). The increase in calcium concentration, due to the addition of lime, allowed the solution to reach saturation with respect to CaUO_4 at pH 8.0. Above this pH, the continued addition of lime caused the continued precipitation of CaUO_4 and the concomitant decrease in uranium concentration.

Figure 3 shows heavy metal concentrations in solution as a function of the pH of the treated groundwater. Only those heavy metals that were affected by the lime treatment are included in Fig. 3.

As with the radionuclides, each metal responded somewhat differently to the lime treatment. The simulated treatment decreased the chromium concentration in groundwater by over an order of magnitude (Table II). The removal of chromium by the precipitation of CrO_2 is analogous to the removal of neptunium and plutonium, since chromium is also multi-valent. In the pH range 7.2 to 7.5 (and Eh range 425 to 400 mV), the concentration of tetravalent chromium (Cr^{+4}) was at a maximum. Consequently, CrO_2 was most stable in this range of pH and Eh (Fig. 3a). The simulated lime treatment decreased the lead concentration in groundwater by almost three orders of magnitude (Table II). The lead concentration was lowest in the pH range 7.1 to 8.1 (Fig. 3a). The precipitation of cerussite (PbCO_3) was responsible for the decrease in lead concentrations. As the pH increased, the carbonate equilibria shifted toward greater concentrations of CO_3^{2-} , allowing the continued precipitation of cerussite. As discussed above, the addition of lime also caused the precipitation of calcite. Above pH 8.1, the continued precipitation of calcite depleted dissolved carbonate concentrations, causing cerussite to redissolve and lead concentrations to increase. Figure 3a shows that a second lead concentration minimum occurred at pH 9.7, caused by the precipitation of $(\text{Pb}(\text{OH})_2)_3 \cdot \text{PbCl}_2$. The groundwater apparently became saturated with this phase as the hydroxide (OH^-) concentration increased. The simulated lime treatment decreased the silver concentration of groundwater by over four orders of magnitude (Table II). Figure 3b shows that the silver concentration remained unchanged until approximately pH 8.1. Above pH 8.1, the silver concentration decrease was linear as a function of pH and was due to the precipitation of native silver (Fig. 3b). The precipitation of native silver was caused by the reduction of dissolved ionic silver, in response to the decrease in Eh that occurred as the simulation proceeded. The mercury concentration decreased slightly at the start of the simulation due to the precipitation of calomel (Hg_2Cl_2). Beginning at

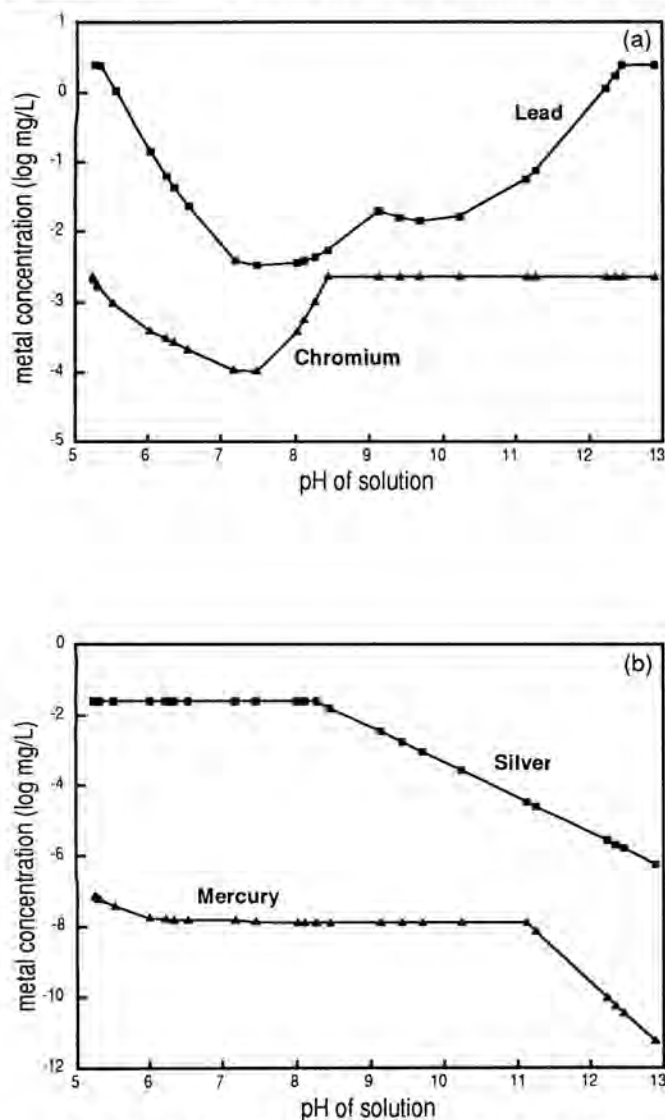


Fig. 3. Metal concentrations versus pH of solution.

approximately pH 11.3, the mercury concentration decreased sharply in response to the precipitation of montroydite (HgO) (Fig. 3b). The precipitation of montroydite was caused by an unexpected increase in the oxygen fugacity of the solution. This increase was an artifact of the simulation, as was the precipitation of montroydite. Thus, although not initially apparent, the simulated lime treatment was not effective in removing mercury from groundwater.

As discussed above, the mass of pure lime required to treat one liter of contaminated groundwater was 1.23 grams. The individual dry masses of the radionuclide- and metal-bearing solids produced during the treatment of one liter of groundwater are shown in Table II. The total dry mass of solids produced during the treatment was approximately 0.57 g L^{-1} of groundwater. This solid waste material was composed primarily of calcite, portlandite, and brucite [$\text{Mg}(\text{OH})_2$]. The mass of lime required for groundwater treatment and the mass of the solid precipitate are dependent on the initial groundwater composition.

CONCLUSIONS

The treatment of contaminated groundwater by pH adjustment was successfully simulated using a standard geo-

chemical code. The simulated treatment required data on the composition of the groundwater, including pH, Eh, and temperature, as input parameters. The following information, specific to the simulation presented in this study, was determined by reaction-path modeling:

- Lime treatment effectively reduced the concentration of americium, neptunium, uranium, chromium, lead, and silver in the hypothetical groundwater. The treatment did not lower the concentration of cesium, strontium, arsenic, or selenium. Although concentrations were slightly lower, the lime treatment was not effective in removing plutonium and mercury from the groundwater.
- Americium and neptunium removal was most complete in the pH range of 9.1 to 9.4, and chromium and lead were most effectively removed in the pH ranges of 7.2 to 7.5 and 7.1 to 8.1, respectively. Uranium and silver removal was optimized at pH 12.9.
- The mass of lime required for treatment was 1.23 g L^{-1} , and the dry mass of solid waste material produced during the groundwater treatment was 0.57 g L^{-1} . The waste material was composed primarily of calcite, brucite, and portlandite.

The simulation indicated that the hypothetical groundwater should undergo batch treatment with lime to optimize the removal radionuclides and heavy metals. Lime should be added until the pH of the groundwater is in the 7.2 to 7.5 range. The solid precipitate should then be removed from the solution by filtration, sedimentation, or some other procedure. The removal of dissolved chromium and lead will be optimized during this first step. Lime should again be added until the groundwater pH is in the 9.1 to 9.4 range. The solid precipitate should again be removed from the solution to optimize the removal of americium and neptunium. The addition of lime should again continue until the pH of the groundwater is approximately 12.9. At this point, further pH adjustment by the addition of lime is not possible. Separation of the precipitate from the groundwater would result in an effluent with the dissolved radionuclide and heavy-metal concentrations listed in Table II. This procedure could be coupled with another process to effectively decrease the concentration of the contaminants that were not attenuated by lime treatment.

The removal of neptunium, plutonium, chromium, mercury, and silver was redox-dependent, and therefore, probably strongly dependent on the choice of performing the simulation as a closed versus opened system. Because of this, the results for these constituents must be interpreted with caution. However, the results for americium, uranium, and lead strongly suggest that these constituents can be effectively removed from groundwater or wastewater using lime to adjust the solution pH.

Based on this study, geochemical modeling techniques are useful in:

- Designing and optimizing laboratory bench-scale tests and screening treatment alternatives, when the contaminants are inorganic, including radionuclides.
- Determining the chemical reactions responsible for the observed behavior of dissolved constituents during the treatment process.

TABLE II
Results Of Simulated Groundwater Treatment

Parameter	Influent Conc. (mg L ⁻¹)	Effluent Conc. (mg L ⁻¹)	Optimal pH Range	Radionuclide- or Metal-Bearing Solid	
				Dry Mass Produced ^a (g L ⁻¹)	Solid Name/Formula
Radionuclides					
americium	40.5	5.46x10 ⁻⁴	9.1-9.4	5.34x10 ⁻²	AmO(HCO ₃)
cesium	1.0	1.0	-	-	-
neptunium	2.08x10 ⁻⁴	9.30x10 ⁻⁹	9.1	2.36x10 ⁻⁷	NpO ₂
plutonium	6.51x10 ⁻¹²	3.88x10 ⁻¹²	5.5-6.0	2.95x10 ⁻¹⁵	PuO ₂
strontium	1.0	1.0	-	-	-
uranium	0.33	5.38x10 ⁻⁸	≥12.9	4.75x10 ⁻⁴	CaUO ₄
Metals					
arsenic	2.5	2.5	-	-	-
chromium	2.29x10 ⁻³	1.05x10 ⁻⁴	7.2-7.5	3.53x10 ⁻⁶	CrO ₂
mercury ^b	7.16x10 ⁻⁸	1.29x10 ⁻⁸	9.1-10.2	6.90x10 ⁻¹¹	Hg ₂ Cl ₂
		5.90x10 ⁻¹²	≥12.9	7.73x10 ⁻¹¹	HgO
lead	2.5	3.41x10 ⁻³	7.1-8.1	3.22x10 ⁻³	PbCO ₃
selenium	1.0	1.0	-	-	-
silver	0.025	5.50x10 ⁻⁷	≥12.9	2.49x10 ⁻⁵	silver (Ag)

^aThe dry mass of the metal-bearing solid precipitated in grams per liter of solution treated.

^bThe precipitation of HgO is an artifact of the simulation. HgO probably would not precipitate.

- Predicting the effectiveness of treatment. Geochemical modeling can predict which contaminants will be removed by treatment and the concentration of the contaminants in the effluent. Geochemical modeling can also be used to predict the chemical conditions required for the optimal removal of contaminants.
- Estimating the mass of chemical reagents required for treatment and the mass of solid waste precipitate generated. Geochemical modeling can predict the mineralogical and elemental composition of the waste precipitate.

The simulation of lime treatment is a single example of the use of geochemical modeling to evaluate and optimize the treatment of contaminated groundwater or a process waste stream. Other applications include the following:

- Simulation of oxidation/reduction processes.
- Analysis of the benefits of pre-treatment (pH adjustment) of water prior to contaminant removal by ion-exchange processes, to ensure that the contaminants are in the optimal form for removal.
- Analysis of the benefits of pre-treatment of water prior to air stripping, to prevent fouling of the strip-per packing by the precipitation of salts.
- Design of waste-processing streams to prevent the formation of scale within critical parts such as heat exchangers.
- Quantification of precipitation and dissolution at points along process streams for the preparation of material balance flow sheets.

Simulations using geochemical modeling techniques can be easily incorporated into feasibility studies and other treatment programs. Treatment simulations can be performed in a timely and cost-effective manner to guide the selection of treatment alternatives and focus bench-scale testing efforts.

REFERENCES

1. P. N. CHEREMISINOFF, "Treating Wastewater," *Pollut. Eng.* 22 (1990).
2. ROCKWELL INTERNATIONAL CORPORATION, "Liquid Waste Treatment Facilities Assessment Report," 87-RF-2571, Rockwell International Corporation, North American Space Operations, Rocky Flats Plant (1987).
3. T. J. WOLERY, "EQ3NR, A Computer Program for Geochemical Aqueous Speciation-Solubility Calculations: User's Guide and Documentation," UCRL-53414, Lawrence Livermore National Laboratory (1983).
4. T. J. WOLERY and S. A. DAVELER, "EQ6, A Computer Program for Reaction-Path Modeling of Aqueous Geochemical Systems: User's Guide and Documentation," Draft, Lawrence Livermore National Laboratory (1989).
5. F. MOREL and J. J. MORGAN, "A Numerical Method for Computing Equilibria in Aqueous Chemical Systems," *Environ. Sci. Technol.* 6 (1972).
6. R. E. McDUFF and F. M. M. MOREL, "Description and Use of the Chemical Equilibrium Program REDEQL2," Tech. Rep. EQ-73-02, Keck Laboratory, California Institute of Technology (1973).
7. J. C. WESTALL, J. L. ZACHARY, and F. M. M. MOREL, "MINEQL, A Computer Program for the Calculation of

- Chemical Equilibrium Composition of Aqueous Systems," Tech. Note 18, Ralph M. Parsons Laboratory for Water Resour. and Environ. Eng., Dept. of Civil Eng., Massachusetts Institute of Technology (1976).
8. J. D. ALLISON, D. S. BROWN, and K. J. NOVOGRADAC, "MINTEQA2/PRODEFA2, A Geochemical Assessment Model for Environmental Systems: Version 3.0 User's Manual," EPA/600/3-91/021, U.S. Environmental Protection Agency, Office of Research and Development (1991).
 9. M. H. REED, "Calculations of Hydrothermal Metasomatism and Ore Deposition in Submarine Volcanic Rocks with Special Reference to the West Shasta District, California," Ph.D. Dissertation, Univ. of California (1977).
 10. M. H. REED, "Calculation of Multicomponent Chemical Equilibria and Reaction Processes in Systems Involving Minerals, Gases, and an Aqueous Phase," *Geochim. Cosmochim. Acta* 46 (1982).
 11. D. L. PARKHURST, D. C. THORSTENSON, and L. N. PLUMMER, "PHREEQE, A Computer Program for Geochemical Calculations," U.S. Geological Survey Water-Resources Investigations 80-96, U.S. Geological Survey, Water Resources Division (1980).
 12. G. H. BRIMHALL, "Deep Hypogenic Oxidation of Porphyry Copper Potassium-Silicate Protore at Butte Montana: A Theoretical Evolution of the Copper Remobilization Hypothesis," *Econ. Geol.* 75 (1980).
 13. G. GARVIN, "The Role of Groundwater Flow in the Genesis of Stratabound Ore Deposits: A Quantitative Analysis," Ph.D. Thesis, Univ. of British Columbia (1982).
 14. J. F. KERRISK, "Reaction-Path Calculations of Groundwater Chemistry and Mineral Formation at Rainier Mesa, Nevada," LA-9912-MS, Los Alamos National Laboratory (1983).
 15. A. E. OGARD and J. F. KERRISK, "Groundwater Chemistry Along Flow Paths Between a Proposed Repository Site and the Accessible Environment," LA-10188-MS, Los Alamos National Laboratory (1984).
 16. D. COLES and L. RAMSPOT, "Migration of Ruthenium-106 in a Nevada Test Site Aquifer: Discrepancy Between Field and Laboratory Results," *Science* 215 (1982).
 17. D. J. ISHERWOOD, "Application of Ruthenium and Technetium Thermodynamic Data Bases Used in the EQ3/6 Geochemical Codes," UCRL-53594, Lawrence Livermore National Laboratory (1984).
 18. D. K. NORDSTROM, "A Comparison of Computerized Chemical Models for Equilibrium Calculations in Aqueous Systems," *Chemical Modeling in Aqueous Systems*, E. A. Jenne, ed., ACS Symp. Ser. 93, American Chemical Society (1979).
 19. T. S. BOWERS, K. L. VON DAMM, and J. M. EDMOND, "Chemical Evolution of Mid-Ocean Ridge Hot Springs," *Geochim. Cosmochim. Acta* 48 (1984).
 20. K. J. JACKSON, "Verification and Validation Studies of the EQ3/6 Brine Model," UCRL-5109C, Lawrence Livermore National Laboratory (1984).
 21. INTERA ENVIRONMENTAL CONSULTANTS, INC., "Geochemical Models Suitable for Performance Assessment of Nuclear Waste Storage: Comparison of PHREEQE and EQ3/EQ6," ONWI-473, prepared for Battelle Memorial Institute, Office of Nuclear Waste Isolation (1983).
 22. C.T. KINCAID and J.R. MOREY, "Geohydrochemical Model for Solute Migration, Volume 2: Preliminary Evaluation of Selected Computer Codes," EPRI EA-3417, Electric Power Research Institute (1984).
 23. J.F. KERRISK, "Chemical Equilibrium Calculations for Aqueous Geothermal Brines," LA-8851-MS, Los Alamos National Laboratory (1981).
 24. U.S. ENVIRONMENTAL PROTECTION AGENCY, "National Primary Drinking Water Regulations," *Code of Federal Regulations*, Title 40, Part 141, Subpart B, U.S. Environmental Protection Agency (1990).