

ENVIRONMENTAL IMPACT OF LANDFILL DISPOSAL OF SELECTED GEOTHERMAL RESIDUES

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ABSTRACT

A solid waste is classified as hazardous if it contains sufficient leachable components to contaminate the groundwater and the environment if disposed in a landfill. Scale, sludge and drilling mud from three geothermal fields (Bulalo, Philippines; Cerro Prieto, Mexico; and Dixie Valley, USA) containing regulated elements at levels above the earth's crustal abundance were studied for their leachability. Cr, As, Cu, Zn and Pb were detected at levels which could impair groundwater quality if leaching occurred. Several procedures were used to assess the likely risk posed by the residues: protocol leaching tests (Canadian LEP and US TCLP), toxicity testing, accelerated weathering test, and a preliminary acid mine drainage potential test. Whole rock analysis, X-ray diffraction, and radioactivity counting were also performed to characterize the samples. Toxi-chromotest and SOS-chromotest results were negative for all samples. Leaching tests indicated that all of them could be classified as nonhazardous wastes. Only one of the six showed a low-level radioactivity based on its high Pb-210 activity. Initial tests for acidification potential gave positive results for three out of six samples while none of the regulated elements were found in the leachate after accelerated weathering experiment for three months.

INTRODUCTION

The exploration and utilization of geothermal resources generate residues such as scale, sludge, and drilling mud. Scale is deposited in steam gathering systems, wellbores, separators, and turbine blades and is manually removed during preventive maintenance shutdown. Sludge is a precipitate of dissolved minerals that forms during cooling of the separated hot brine at the sludge sump or holding ponds prior to disposal. Drilling mud is a by-product of drilling operations during the exploration and development of

the geothermal sources. Most geothermal residues have posed disposal problems to geothermal operators since some have been reported to contain toxic elements at elevated concentrations above normal soils (Hickman, 1988; Gallup and Reiff, 1991; Peralta et al, 1996). Landfilling is widely practiced as a disposal option since it is inexpensive and perceived to have low environmental impact.

Environmental impact assessment (EIA) for a particular activity or project has traditionally been focused on evaluating the effects on the various sectors of the environment such as on air, land and water. Thereafter through weighting or valuation techniques, a summary of predicted impacts is presented in a matrix along with the proposed mitigating measures. The basis for arriving at most predictions are not yet refined sufficiently to be objective hence most of these so-called probable impacts are still basically inaccurate. Elimination of this uncertainty aspect in impact assessment requires more evidence to support predictions especially the environmentally significant ones. The scoping process, which is a first level screening method, can immediately rule out minor impacts to direct attention to important issues related to the activity. In scoping for this study, the likely impacts of landfill disposal of geothermal residues on groundwater were found to be of significance compared to the impacts on other environmental media.

To many decision-makers, prediction is the key component of an EIA methodology since it can have a more rigorous approach and objective assessment. Modes of analysis may be in the form of instrumental techniques and through laboratory and computer simulation studies. In disposing geothermal residues in a landfill, the most serious threat to the environment is leaching of the toxic components (such as heavy metals) to groundwater which eventually affects human population. Further

investigation of conditions and mechanisms under which metals might be eventually released would clarify any potential environmental contamination. If the samples are found to be nontoxic and leaching is insignificant, then these particular residues from the three geothermal fields are suitable for landfilling or land reclamation with minimal or no treatment.

Waste characterization, as an emerging tool in environmental engineering in identifying the true nature of particles, has been used as a first step screening. It involves determination of particle size, chemical content, dominant phases, and radioactivity. Second level studies are leachability and toxicity testing. Protocol leaching procedures to categorize wastes as hazardous or nonhazardous as well as to predict their long-term leaching behavior have been widely used. Leaching can be enhanced by natural weathering over time or by certain extreme conditions. The accelerated weathering test and acid mine drainage test can be used to ascertain this susceptibility.

Numerous protocol leaching tests are available but the most widely used are Toxicity Characteristic Leaching Procedure (TCLP) in the US (USEPA, 1990) and Leachate Extraction Procedure (LEP) in Canada (Government of Ontario, 1990). Both procedures assume co-disposal of municipal wastes and industrial wastes in a sanitary landfill. Both methods use a 20:1 liquid to solid ratio, the rotary extraction mechanism and a buffered extracting solution. Main differences are that the Canadian LEP is two hours longer with intermediate pH adjustments and extraction at slower rotation speed (10 rpm against the 30 rpm of TCLP). These procedures are described more in detail under experimental work.

Two microbial colorimetric bioassays have been recently developed, SOS-Chromotest and Toxi-Chromotest to detect genotoxic and toxic activities of chemicals, pharmaceuticals, and food stuffs. The tests have also been applied to environmental samples such as water, sewage and sediments (Xu and Dutka, 1987). The toxicity bioassay is based on the ability of toxicants to inhibit the de novo synthesis of an inducible enzyme, β -galactosidase. The amount of de novo synthesized enzyme is determined by a colorimetric reaction.

The SOS-Chromotest is a qualitative but semi-quantitative method of detecting the presence of genotoxicants (Quillardet and Hofnung, 1985; 1993). A genotoxicant or genotoxin is any DNA-damaging agent, e.g., mutagen which attack the genome (DNA) part of the cell. A genetically engineered strain of *E.*

coli PQ37 (Institut Pasteur, France) was developed to mimic an organism's response to genotoxins. Genotoxins affect living cells by altering or creating lesions in the DNA structure causing mutations through faulting base pairing during the excision and repair pathway. The effect of toxins on living cells is more rapid and simply causes cell death. On the other hand, the Toxi-Chromotest involves a different strain of lyophilized *E. coli* K12 OR85 bacteria (Organics, Israel) produced by arbitrary bombardment with UV light. This bioassay is used to measure the ability of the toxin to inhibit enzyme production by killing the organism.

This series of tests can provide data to reliably predict on a microscale level the major environmental impact of landfill disposal of selected geothermal residues. The objectives of this study are (a) to determine the risk involved if geothermal residues were disposed in a landfill, (b) to study their possible leaching behavior under various protocol tests, (c) to investigate their potential toxicity, and (d) to examine long-term behavior as a result of weathering or acidification.

EXPERIMENTAL PROCEDURES

The six geothermal residues were obtained from three geothermal fields: (a) Bulalo, Philippines, (b) Dixie Valley, USA, and (c) Cerro Prieto, Mexico. They were all examined on an as-received basis since they were relatively dry with an average moisture content of less than 5%. Each sample was assigned the following codes to facilitate the discussion: PSC - Philippine scale, PSL - Philippine sludge, ASC - American scale, MSC - Mexican scale, MSL - Mexican sludge, and MDM - Mexican drilling mud. PSC, PSL, MSL, and MDM have fine or flaky particles below 9.5 mm in size while MSC and ASC are hard and rock-like composed mostly of big particles ranging from 1 to 15 cm with a fraction of fine to coarse particles.

Whole Rock Analysis

Approximately 10 g each of the six air-dried geothermal samples was analyzed for whole rock analysis, which includes 32 elements including sulfur and mercury. The sulfur content is useful in the calculation of acid production potential which will be described later.

X-Ray Diffraction

Powder X-ray diffraction (XRD) was performed on the samples to identify the mineral phases present and determine their crystal structures. A Siemens D5000

diffractometer system using $\text{CuK}\alpha$ ($\lambda = 1.54178\text{\AA}$) radiation was used in scanning from 5° to 65° 2θ . Phase identification was carried out using 1989 Hanawalt Index of the Joint Committee on Powder Diffraction Standards JCPDS/PDF-2 Data Set.

Radioactivity Counting

Radioactivity of about 5 g per sample was measured using a hyperpure germanium (HPGe) well-type detector. Measuring time for all samples was between 10,000 to 82,000 seconds using as reference standard a soil sample with known values of U, Ra, and daughter's activities. Since MSC showed unusual radioactivity levels, four confirmation runs were carried out with measuring time between 70,000 to 413,000 seconds (1 to 5 days). Spectrum analysis and activity calculations were derived using equations from 1991 APTEC OSQ/Plus Manual.

Toxicity Testing

The samples for toxicity tests have been prepared using two kinds of solid extraction methods: (a) 10% dimethylsulfoxide (DMSO) + 10% methanol and (b) direct sediment testing procedure (DSTP) (Kwan, 1995). Both bacteria, *E. coli* strain PQ37 and *E. coli* K12 OR85 were obtained from Environmental Bio-Detection Products, Inc., Brampton, Ontario.

The SOS-Chromotest was performed using 100 μL of an exponential growth phase culture of *E. coli* strain PQ37 into all the wells in a standard 96 well microtitration plate. Following a two-hour incubation at 37°C , 100 μL of blue chromagen was added to the wells and reincubated for another hour. Genotoxic activity is noted by the presence of a distinctive blue colour in the wells. A relative measure of genotoxicity is determined by measuring the intensity of the blue color using a spectrophotometer.

For the Toxi-Chromotest, serial two-fold dilutions of the samples were prepared in the microplate. A vial of *E. coli* strain K12 OR85 was rehydrated and mixed with the reaction mixture. Around 100 μL of this mixture were added to each well in the microplate. Following a 90 min incubation at 37°C , blue chromagen was added and reincubated for another 90 min. Toxic activity is noted by the absence of blue color (Kwan and Dutka, 1992).

Leachate Extraction Procedure (LEP)

A 50 g dry sample (9.5 mm or less particle size) was tumbled continuously in a 1 L polyethylene bottle containing 800 mL of deionized water for 24 hours in

a rotary extractor at 10 rpm and at room temperature. A pH of 5 ± 0.2 was maintained throughout the extraction by adding 0.5N acetic acid at 1, 3, 6, 22 hrs from starting time. No more than 200 mL of acetic acid may be added. After completion of the extraction, the slurry was centrifuged and the supernatant leachate was filtered through 0.45 μm cellulose acetate filter paper. The leachate was analyzed for the elements of interest using inductively coupled plasma emission spectrometry.

In addition, the effect of particle size on metal leaching was investigated by carrying out the LEP test on MSC using three different particle sizes : <125 μm , <4 mm, and 6-9.5 mm. MSC was of particular interest since chemical analysis showed it had the highest levels of Pb, Zn, and Cu among the six samples.

Toxicity Characteristic Leaching Procedure (TCLP)

Similar to the LEP, the TCLP maintains a 20:1 liquid to solid ratio with particle size 9.5 mm or less. Extraction was carried out continuously in a rotary extractor at 30 rpm rotation for 18 hours. The choice of extraction fluid was dependent on the initial pH of the sample (taken after 5 minutes of magnetic stirring in deionized water). If the pH is <5, extraction fluid #1, composed of a buffer at pH 4.7 of dilute acetic acid and 1N NaOH, was used. On the other hand, if the pH is >5, extraction fluid #2, composed mainly of dilute acetic acid (pH 2.8) was used instead. After the 18-hour extraction, the supernatant was filtered with a 0.45 μm membrane filter and analyzed for various elements using inductively coupled plasma emission spectrometry.

Likewise as in LEP, the effect of particle size on metal leaching was studied by using the TCLP test on MSC using three different particle sizes: <125 μm , <4 mm, and 6-9.5 mm.

Preliminary Acid Mine Drainage Potential Test

The B.C. Research Initial Test (BCRIT) was used to measure acid-consuming and acid-producing components of the residues (CANMET, 1991). In a 250 mL beaker containing 100 mL of deionized water, a 10 g pulverized sample (100 mesh) was stirred continuously with a magnetic stirrer. The natural pH was measured after 15 minutes. While stirring, the sample slurry was titrated with 1N H_2SO_4 to an endpoint of pH 3.5. The volume of acid consumed was noted and used for calculation of acid production potential. The sulfur content of the sample must be known from the chemical analysis to

be used in estimating the acid potential of the sample.

Accelerated Weathering Test

In a 250 mL Erlenmeyer flask containing 70 mL deionized water and bacteria nutrient media, a 10 g pulverized sample (100 mesh) was shaken continuously inside an incubator-shaker for 3 months at 150 rpm and 35 °C. The pH was monitored weekly and the flasks were weighed and deionized water was added to make up for weight loss due to evaporation. At the end of the shaking, 15 mL aliquot was obtained for metal analysis (CANMET, 1991).

RESULTS AND DISCUSSION

Waste Characterization

Chemical analysis of the six geothermal residues revealed that they are composed predominantly of silica ranging from 66-82% by weight. PSC and PSL contain high levels of iron and aluminum content (4-8%) followed by MSC and MDM (2-4%). ASC is essentially an aluminosilicate due to its high levels of alumina(10%) and silica (67%) with all the rest of the elements in trace quantities. MSL, with its fine white silty particles, is composed mainly of silica (82%) with little contamination. From the chemical analyses, it was apparent that ASC and MSL will be less of a concern while PSC, MSC, PSL and MDM contain above normal crustal levels of Pb, Zn, Cu, Cr, As, Mn and Ba. In particular, MSC is concentrated with Cu, Zn, and Pb (around 1% each). Both PSC and PSL have the most As content of 300 ppm. MSC and PSC have S content typical of mine tailings.

X-Ray Diffraction

Several dominant phases were detected by XRD in the samples as shown in Table 1. Almost all of the samples, except MDM, contain an amorphous silicate phase with a broad maximum at around 4 Å. ASC does not contain any crystalline material but only pure amorphous silicate, possibly aluminosilicate as indicated by whole rock analysis. MSL is mainly halite and sylvite while MSC contains galena, sphalerite, chalcopryrite, and cubanite. Both PSL and PSC contain quartz, magnetite, and hematite. MDM is the most complex containing quartz, calcite, pyrite, halite, feldspars, clay and silicates. Knowing these phases will be useful in predicting the weatherability of the residues. In terms of their possible environmental impacts, the natural minerals and layer silicates are relatively inert while halite can dissolve and sulfides may oxidize releasing heavy metals.

Table 1. X-Ray Diffraction Data of Selected Geothermal Residues

Sample	Species*
ASC	AMM at 3.43 Å
PSC	AMM at 4.10 Å Quartz Magnetite Hematite Jarosite
MSC	Galena Sphalerite Chalcopryrite Cubanite AMM at 3.80 Å
PSL	Albite Hornblende Quartz Hematite AMM at 4.04 Å
MSL	Halite Sylvite AMM at 4.04 Å
MDM	Quartz Calcite Halite Albite Microcline

*Listed in order of abundance
AMM - Amorphous material maximum

Radioactivity Counting

The radionuclides detected were Th-230, Pb-210, Ra-226, Ac-228, K-40 and total U. The most important radionuclide to monitor is Ra-226 since it decays to radon which is toxic when inhaled. All the activities are in the range of NORM (naturally occurring radioactive materials) with the exception of Pb-210 ($t_{1/2} = 22$ y) in the MSC sample. The validation counting for MSC at longer duration (up to 5 days) gave an average measurement of 130,000 Bq/kg (3513 pCi/g) for Pb-210 at 90% confidence level. The amount of radioactive Pb-210 in the MSC sample was found to be insignificant at 45×10^{-6} ug/g which is only $4 \times 10^{-7}\%$ of the total Pb (11,600 ug/g). Gallup and Featherstone (1995) reported 250-400 pCi/g in the Salton Sea geothermal brines in

southeastern California where the anticipated NORM regulation for solid wastes is 5 pCi/g. The Pb-210 activity of 130,000 Bq/kg is comparable to a radiation dose of 32.5 mSv/y (received via ingestion) or 14 times the total annual effective dose equivalent from all natural sources of 2.4 mSv but which is still lower than the current occupational dose limit of 50 mSv/y (UNSCEAR, 1993). There are regions in the world where outdoor terrestrial background radiation levels appreciably exceed the NORM at 2-6 times the average natural background of 1 mSv/y : Guarapari, Brazil; Kerala, India; and Yanjiang County, Guangdong, People's Republic of China. This is due to the presence of monazite sands with high levels of thorium, uranium and radium. The inhabitants in these areas were studied between 1970-1985 and there was no increase in the frequency of cancer among the population (ICRP, 1991; UNSCEAR, 1993).

Toxicity Testing

These two tests provide an indication of the sensitivity of bacteria to toxic elements. Toxi-Chromotest and SOS-Chromotest results were negative for all samples. However, negative results may also indicate that the genotoxicants may be bound within the residue matrix or are insoluble in either water or the extractant. This was confirmed by the leaching tests and the XRD which identified that some toxic elements are present as insoluble sulfides or silicates. Therefore as long as the samples are disposed or maintained in this stable or inert state, they are not considered genotoxic.

Protocol Leaching Tests

With the protocol mesh screening of 9.5 mm, all the samples pass the LEP and TCLP, i.e., the regulated elements were not detected at registrable levels in the leachate. However, at a worst case scenario, using a finer particle size of <125 μm , Pb in MSC was found to leach out above regulatory limits. As shown in Figure 1 for LEP, with increased surface area and the exposure of galena to oxidation during pH adjustments, around 0.1% of the original Pb was dissolved and found in the MSC leachate (12 ppm against the limit of 5 ppm). In Figure 2 for TCLP, MSC particles with <4 mm in size are likely to leach Pb at twice the limit and at <125 μm will release 20 times more. In comparing the two procedures, TCLP appears to be a more aggressive procedure than LEP which could be due to its higher rotation speed. This means that although the elements exist in the residues, the samples can be classified as nonhazardous wastes at 9.5 mm size since the heavy metals are in insoluble forms as shown by the XRD or may be trapped in the

silicate matrix and are unable to leach unless ground to powder size. The residues can therefore be disposed in a secure landfill at bulk sizes while the fines must be stabilized or treated appropriately prior to disposal. Existing treatment methods for geothermal residues are biotechnology and solidification (Webster and Kukacka, 1987; Peralta, et al., 1992; Premuzic, 1993).

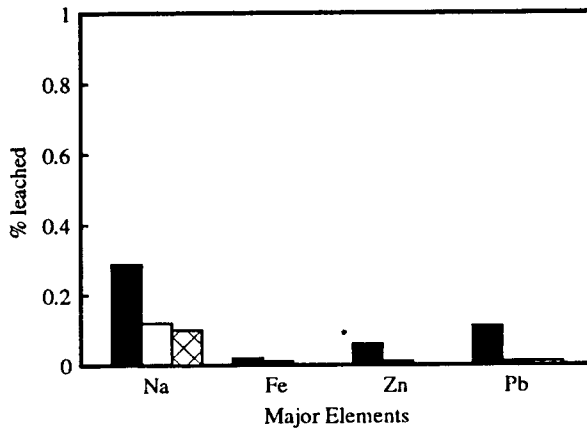


Fig.1. Effect of particle size on leaching (LEP) : MSC

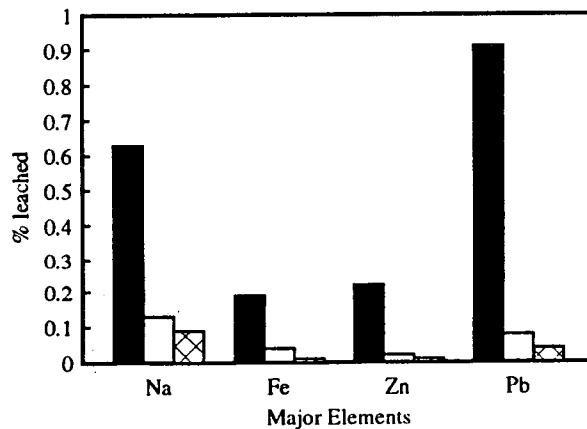


Fig.2. Effect of particle size on leaching (TCLP) : MSC

■ <125 μm □ <4 mm ☒ 6-9.5 mm

Preliminary Acid Mine Drainage Potential Test

The B.C. Research Initial Test (BCRIT) determines the neutralization capacity of the sample and its acid producing potential. In comparing the two values, if

the acid production potential (APP) exceeds the acid consumption (AC) expressed in kg H₂SO₄ per ton of material, the sample is classified as potential acid producer and confirmation testing is recommended. This corresponds with the acidity contribution from H₂SO₄ as a result of its formation due to oxidation of the sulfides to sulfates. The acid production potential was calculated from the percent sulfur in the sample converted to kg H₂SO₄ whereas the acid consumption was computed from the volume of acid used to reach the endpoint of pH 3.5. As shown in Table 2, only three samples, PSC, MSC, and PSL have been found to have acid mine drainage potential (positive values) which would require verification. The choice of the endpoint of pH 3.5 is based on the assumption that this represents the limit above which iron and sulfur oxidizing bacteria such as *Thiobacillus ferrooxidans* are no longer active. Therefore, if the theoretical acid production is not sufficient to lower the pH to below 3.5, then biochemical oxidation of the wastes will not occur and the formation of acid mine drainage is unlikely (CANMET, 1991).

Table 2. Initial acid mine drainage potential test

Sample	pH	% S	APP	AC	APP-AC
PSC	3.5	1.68	51.4	0.98	+50.42
MSC	7.2	3.38	103.43	5.39	+98.04
PSL	4.5	0.46	14.07	0.98	+13.09
ASC	9.7	0.03	0.92	6.86	-5.94
MSL	7.8	0.01	0.31	2.94	-2.63
MDM	8.3	0.1	3.06	86.24	-83.18

APP - acid production potential, kg H₂SO₄

AC - acid consumption, kg H₂SO₄

Accelerated Weathering Test

The accelerated weathering experiments provided further insights into long-term behavior of geothermal residues in a landfill environment. Chemical weathering is related to such factors as climate, topography, parent material, and time with temperature and moisture flux as the major environmental variables affecting weathering rates (Birkeland, 1974; Bohn et al., 1985). From the XRD data (some not listed on Table 1), the easily weatherable minerals are halite, gypsum, sulfides,

calcite, dolomite, hornblende, primary layer silicates and albite. Around 5% of the original amount of Al from its silicates and 30% of the transition metals Mn, Fe, Cu and Zn (probably from their sulfides) were found in the leachate after three months of continuous shaking. None of the regulated elements (As, Cr, Cd, Ba, and Pb) was detected in the leachate.

CONCLUSIONS AND RECOMMENDATIONS

All the geothermal residues tested do not contain radioactive materials and genotoxicants. They are not expected to contaminate the groundwater if disposed in a landfill. MSC, PSC, and PSL have been found to have acid mine drainage potential and it is proposed to conduct confirmation tests to obtain kinetic information on metal leaching under biochemical oxidation. In addition, computer simulation of leaching can be undertaken using geochemical models (such as MINTEQA2) to determine thermodynamic reactions and predict equilibrium concentrations. It is unlikely that the fine-sized Mexican scale will leach out unless subjected to extreme conditions such as acidic solution and constant weathering. As a precaution however, the smaller particles of <4 mm must be stabilized prior to disposal.

The predictions made in this work are true for the particular samples from the three geothermal fields. It is probable that similar behavior can be inferred from other geothermal residues from other sites having similar characteristics.

ACKNOWLEDGMENT

The authors wish to thank the Philippine Geothermal Inc., Comision Federal De Electricidad (Proyectos Geotermoelectricos) of Mexico, and Oxbow Geothermal Inc., Nevada for providing the samples for this study. We also acknowledge the assistance from colleagues at the University of Toronto : Ms. Sheree Yiin, Department of Microbiology for the toxicity tests; Prof. Greg Evans and Dr. Sandu Sonoc, Center for Nuclear Engineering, for radioactivity counting and interpretation; Dr. Kishor Mehta and Ms. Jane Y. Gerardo, International Atomic Energy Agency, Vienna and Dr. Marcelo Lippmann of the Lawrence Berkeley Laboratory, California for their comments, advice and continuous support.

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