

**Potential Health Impact of Complex Chemical and Radionuclide Mixtures  
Due to Proposed Nuclear Waste Repositories**

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**ABSTRACT**

The U.S. Department of Energy (DOE) is proceeding with its license application to the Nuclear Regulatory Commission to approve the proposed Yucca Mountain Repository. The authors have identified several areas where additional scientific studies are needed and should be investigated by the DOE prior to submission of the license application. First, there are concerns associated with the corrosion of the metal canisters and the release rates of radionuclides and heavy metals into the environment. Second, there are questions regarding uncertainties associated with the use of the Total System Performance Assessment Model (TSPA) employed by the DOE. Third, the Performance Assessment Model did not consider the competing effects of sorption of radionuclides and heavy metal mixtures. There is a need for additional experimental work in this area. Finally, the potential toxicological interaction between radionuclides and heavy metals must be further studied.

**KEY WORDS:** Risk assessment; radionuclide; heavy metals; complex mixtures, sorption; interaction; toxicity; and synergism.

**I. INTRODUCTION**

The U. S. Department of Energy (DOE) is planning to submit a license application for the Yucca Mountain Site to the Nuclear Regulatory Commission by December 2004. The authors have identified several areas where additional scientific data need to be obtained by the DOE prior to license application. It appears that there are several major uncertainties and scientific data deficiencies that must be addressed by additional research before the Yucca Mountain Repository can be approved as the nation's first high-level nuclear waste repository. This conclusion is in contrast to that expressed in a recent Nature commentary by Boyle (1). He stated that the only real concern was possible future volcanic activity in the Yucca Mountain region. In the current paper, several scientific issues and uncertainties associated with approval of the Yucca Mountain site as a high-level nuclear waste repository are discussed.

**II. METAL CORROSION**

A major concern at Yucca Mountain will ultimately be the health risk to human populations in the future because of canister failure and the subsequent migration of radionuclides and heavy metals into the groundwater. A potential scenario for groundwater contamination with chemicals and radionuclides from the repository is as follows. First, a plume of heavy metals including Cr, Co, Ni and Ti will be generated from the corrosion of the spent fuel canisters and the drip shields. Next, dissolution of the fuel waste forms will result in the release of radionuclides including suspected carcinogens, such as gadolinium. Finally, long half-lived radionuclides, such as Tc-

99, I-129, Pu-239, and Np-237, are expected to migrate from the site into the accessible environment over time (2).

The health risk posed by releases of mixtures of toxic metals and radionuclides into the environment because of corrosion of the canisters and subsequent failure of the engineered barriers remains unknown and has not been completely investigated. There has been debate within the scientific community about possible contributions from the so-called "protective layer" that forms as a result of the corrosion of alloy C-22, which has been proposed to be used in the spent nuclear fuel canisters, and Ti-7 to be used for the drip shields (3). For example, the protective layer could break down due to physical mechanisms such as mechanical abrasion, rock falls, and seismic activity.

At least one study by the state of Nevada has shown that these metals canisters could be corroded under various laboratory conditions (4). In addition, Nevada recently reported that corrosion rates vary from 0.1 mm to 1.0 mm per year, and may reach a peak value of 10 mm per year (5). Microorganisms could also contribute to the accelerated corrosion of canisters. This problem area has not been fully investigated (6). Moreover, microorganisms may enhance migration of heavy metals from the proposed repository. In the Final Environmental Impact Statement (FEIS) for the Yucca Mountain Project (7), the composition and the amount of various substances to be buried included 86,000 tons of alloy 22 containing 22.5% Cr, 14.5% Mo, 57.2% Ni and 0.35% V; along with 140,000 tons of stainless steel is 17% Cr, 12% Ni, and 2.5% Mo. The health risks posed by the potential release of a fraction of this amount of heavy metals along with radionuclides must be further addressed.

### **III. NUCLIDE SORPTION AND THE TOTAL PERFORMANCE ASSESSMENT**

#### **A. Total System Performance Assessment**

In order to assess the public health risk associated with the behavior of radionuclides in the environment, knowledge of the partitioning of each radionuclide between different phases is required. This requires information on the basic physicochemical properties of the radionuclides, soil/mineral surfaces, and colloids/particulates and dissolved complexes. A distribution coefficient ( $K_d$  value) describes the partitioning of a radionuclide between the solid and aqueous phase of a system and ultimately provides an estimate of each radionuclide's transport interactions and movement via the groundwater pathway.

When modeling sorption, the YMP Performance Assessment did not consider competing effects of radionuclides and heavy metals. While sorption properties of individual radionuclides and heavy metals may be known (mostly in the near field), variations in these properties when two or more radionuclides and heavy metals are present is not. For instance, a canister must degrade before the radionuclides can be released. Therefore, heavy metals such as Ni, Cd, Cr, Ti, and Mo will migrate from the site first and be adsorbed within the near field. This limits the number of soil binding sites for subsequent radionuclide sorption. Furthermore, the EIS states that sorption parameters measured for a single radionuclide are applicable to the case where more than one radionuclide is present. Competitive effects are assumed to be negligible. This requires confirmation for near-field conditions.

In a letter dated January 17, 2002, the Chair of the Advisory Committee on Nuclear Waste write to the Chair of the U.S. Nuclear Regulatory Commission (8) and stated the following: "The Committee believes that risk-informed regulatory decision making should be conservative, but be based on realistic and reasonable analyses. A risk-informed performance assessment should be a realistic representation of the risk, including quantification and importance ranking of the sources of uncertainty. That is, the performance assessment should represent the best attempt of the experts at quantifying the risk, and it should not be obscured by assumption-based conservatism. The use of the assumption-based conservative analysis for performance

assessment compromises the regulator's ability to quantify defensible safety margins. Assumption-based modeling conservatisms can be wrong in both the likelihood and consequences of events and may not result in the best risk-informed regulatory decision making."

Furthermore, the joint NEA-IAEA International peer review of the YMP's TSPA (9) included the following "It is recommended that more experimental data be obtained to build confidence in the thermodynamic modelling, especially with regard to the complex interactions between the waste form and components of the waste package."

In a recent publication by S. Nadis in Scientific American (10) he cited that Rodney C. Ewing, a member of the National Academy of Science Waste Management Board on Radioactivity and a member of the Yucca Mountain Peer Review Panel. Ewing raised several scientific concerns challenging the adequacy and uncertainties associated with the use of the TSPA by DOE at YMP such as: "We've learned a lot about the mountain, but when we look at the substance of it, our knowledge is actually quite thin." Additionally, in his opinion, "the mathematical approach keeps us from seeing how the individual components are working. For example, much stock is being placed in alloy 22, a relatively untested metal that is supposed to confine waste over the long haul."

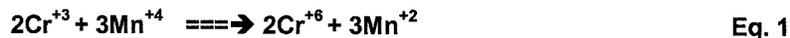
Uncertainties throughout the model are rolled together, makes it hard to tell whether any of the barriers are effective. He also added, "There's been no attempt to test this module on real geological systems." Computer simulation models of YMP release rates and migration rates of heavy metals and radionuclides into the environment may introduce serious scientific errors.

Risk assessment associated with complex mixtures constitutes a critical element of the total system performance (TSPA) model for the Yucca Mountain High-Level Nuclear Waste Repository. Since the TSPA must predict what will happen at the YMP site over 10,000 years and what risk to humans may result, it is not possible to validate the TSPA model in advance. However, it is possible to apply appropriate Quality Assurance/Quality Control (QA/QC) measures to predict risk levels within the TSPA model. An important part of the QA/QC work should include a sensitivity and uncertainty analysis for all input data regarding complex mixtures.

## Chromium Toxicity and Cancer Concerns

The Yucca Mountain Project concluded that corrosion of canisters will promote the formation of silicates due to contact with the host rock. This will precipitate most of the Ni and Mg released from the canister walls, but virtually none of the Cr, Mo, and V will precipitate. However, chromium is a highly toxic and carcinogenic species when in oxidation state Cr<sup>+6</sup>. It is also highly mobile in an alkaline environment, such as Yucca Mountain. Cr<sup>+3</sup> is less toxic and is not a carcinogen.

Palmer and Puls (11) stated that any evaluation of the natural attenuation of Cr<sup>+6</sup> must consider the potential oxidation of Cr<sup>+3</sup> to Cr<sup>+6</sup>, Equation 1 illustrates the oxidation of Cr<sup>+3</sup> by MnO<sub>2</sub>.



Based on a review of the YMP draft EIS (2), it appears that the oxidation rate of Cr<sup>+3</sup> to Cr<sup>+6</sup> by manganese dioxide in the unsaturated and/or saturated zones was not completely investigated or modeled. The presence of manganese dioxide in large quantities at various locations within the proposed repository has been documented. Cr<sup>+6</sup> is soluble over a wide range of pH, and highly mobile in most natural environments including alkaline soils as described by Khan and Plus (12).

#### IV. ASSESSMENT OF COMPLEX MIXTURES

The adverse health effects from exposure to mixtures of heavy metals and radionuclides have not been addressed in the Final Environmental Impact Statement (FEIS) (7). The effect of free radicals produced in cells by ionizing radiation may be intensified and by environmental co-pollutants for example (e.g. heavy metals). Heavy metals such as Cr have been known to produce reactive oxygen species (ROS). Increased concentrations consequently leads to ROS stress and depleted of the normal cell antioxidant defense capacity. This subsequently leads to increases in lipid peroxidation (LPO) that can induce DNA damage, produce mutagenesis, apoptosis, and possibly cancer.

In 1986, the U.S. EPA issued guidelines for the risk assessment of complex mixtures (13), and subsequently issued the 1989 Risk Assessment Guideline for Superfund (14). In 1990 the EPA published a technical Support Document to provide more detailed information on toxicity of whole mixtures and toxicological interactions (e.g., synergism) between chemicals (15). In 2000 the EPA issued a new guideline (16) for complex mixtures entitled "Supplementary Guidance for Conducting Health Risk Assessment of Chemical Mixtures." In 2001, the Nuclear Regulatory Commission (NRC) published guidelines and recommendations for dealing with exposure to multiple radionuclides (17).

The Agency for Toxic Substances and Disease Registry (ASTDR) (18) in 2001 issued draft guidelines for chemical interactions. Additionally, the Agency for Toxic Substances and Disease Registry is currently drafting proposed guidelines for chemical interactions and radionuclide interactions such as; Ce, Co, Sr, and Trichloroethylene (19). On March 31, 1995 the Office of Environmental Policy and Assistance within the U.S. Department of Energy issued a memorandum (20) and guidance document entitled "CERCLA Baseline Risk Assessment Reference Manual for Toxicity & Exposure Assessment and Risk Characterization." This memorandum and guidance set a general framework on how DOE should; (1) require the contractor personnel to take into consideration EPA guidance, and, (2) help project personnel to discuss EPA guidelines with regulators, decision makers, and stakeholders as it is related to conditions at a particular DOE environmental restoration site. The National Research Council (NRC) in 1988 also addressed concerns regarding exposures to complex mixtures (21). The Presidential/Congressional Commission on Risk Assessment and Risk Management in 1977 stated that it "considered the risk assessment of mixtures to be a matter of considerable concern and importance (22)."

Very little research is currently available on the potential interactions between chemical agents and radiation. There are examples of important interactions between chemical agents (e.g. asbestos and smoking) and a few reports that document interactions between chemical agents and radiation (e.g. combined chemotherapy and radiation therapy increases the risk of secondary leukemia). A discussion of chemical risk from a nuclear repository in Canada by Goodwin et al., (23) in 1987 pointed out a significant risk for man and the environment from a nuclear waste repository. The authors recommended that an assessment of potential chemical impacts along with radiation should be part of the formal safety assessment. In 1993, the National Council on Radiation Protection and Measurements (NCRP) (24) specifically acknowledged that a gap exists between chemical and radiation risk estimates. In addition, the NCRP confirmed that further study is needed to address issues such as damage to the immune system, and the possible combined effects of chemicals and irradiation causing either synergistic or antagonistic effects.

Persson (25) in 1990 recommended that part of the safety analysis, can be used for a nuclear waste repository, should include evaluation of chemo-toxic impacts on man and the environment. He also pointed out, that for the short, term the radiological toxicity of materials in a nuclear waste repository is of primary concern. However, the potential chemical toxicity should not be overlooked. For the longer time frame of a million years, as radioactive materials decay, chemical toxicity may even become more dominant. In 1995 Forme (26) developed a statistical methodology can be used for designing experiments and analyzing data related to health effects

(including mutagenicity and carcinogenicity) of chemicals and irradiation. This research emphasizes the design and analysis of biological experiments for determination of dose-effect relationships

Over the past several years efforts have been made to develop methodologies for risk assessment of chemical mixtures, but mixed exposures to two or more dissimilar agents such as radiation and one or more chemical agents have not yet been addressed in any substantive way. In 2000, the United Nation Scientific Committee on the effects of atomic radiation (27) issued an extensive report on the combined effects of radiation and other agents. It was concluded that a comprehensive approach for the study and quantitative assessment of combined effects of radiation and chemical agents must be developed. The gap between different conceptual approaches in the assessment of risks associated with chemical toxicology and radiological protection has to be bridged. Multidisciplinary approaches to this research have to be forged.

Chen and McKone (28), under a DOE sponsored grant, conducted an extensive literature review concerning the health effects associated with exposure to ionizing radiation and chemicals. The authors concluded that very little quantitative information is currently available on the cumulative effects of exposure to multiple hazardous agents that have either similar or different mechanisms of action. Their literature review summarized three types of interactions between radiation and chemicals.

- 1) synergistic effects, exemplified by the observation that exposure of human lymphocytes to benzene and radiation showed an increase in chromosome aberrations and concurrent smoking and radon exposures in uranium miners resulted in an increase in the lung cancer rate. In both of these cases, the combined effects of exposure were greater than the sum of the effects caused by each agent acting individually.
- 2) also, additive effects such as those resulting from smoking and ionizing radiation exposure in Japanese atomic bomb survivors.
- 3) antagonistic effects exemplified by *in vitro* exposure of rodent cells to ionizing radiation and selenium resulted in fewer radiation-induced cell transformations produced than observed with radiation alone (Chen and McKone, 2001).

Nosove et al., (29) reported on the effects of exposure of rats to a mixture of two radionuclides (Cs-137 and Sr-85) for exposure over 30, 56 and 90 days. Test results showed ultrastructural changes of lipid peroxidation in rat cerebrum and blood. Maximum biochemical changes have been detected after radionuclide administration for 30 days. They noted a spontaneous increase in chemiluminescence level. Evaluation of the experimental data strongly suggest that mixed radiation increased levels of ROS induced ROS stress and enhanced synergistic interactions as seen in the increased levels of LPO which resulted from exposure to the two radionuclides, Cs-137 and Sr-85.

ATSDR (19) cited several Russian epidemiological studies illustrated an interaction between Sr-90 and Cs-137 in a population in the Ural Mountains undergoing chronic exposure. The estimated dose levels for the exposed population were 3-4 sieverts (Sv) from 1949 –1956, 0.9 Sv for the 1957 release, and 0.003 Sv for the 1967 release. The study demonstrated that the exposed population had a variety of medical symptoms, such as: chronic radiation sickness characterized by hematological symptoms, neurological disturbances, immune system changes, and cardiovascular changes. A significant increase in long-term morbidity and mortality, both general and cancer specific, were also observed.

An example of the interaction between UV-radiation and nickel was reported by Lynn et al., (30). They investigated the effect on inhibition of DNA repair in Chinese hamster ovary cells. Nickel has been known to inhibit DNA repair, and nickel also can increase cellular reactive oxygen species in cells. Test results demonstrated the generation of hydrogen peroxide free radicals in the presence of nickel and exhibited a synergistic inhibition on both DNA

polymerization and ligation, can cause protein fragmentation. Also, nickel can induce irreversible damage to the proteins involved in DNA repair, replication, recombination, and transcription.

In light of all of the studies and scientific evidence available in the literature over the past several years, efforts have been made to develop methodologies for risk assessment due to exposure to chemical and radionuclide mixtures. The fact is that there are Federal guidelines already in place for estimating such effects from such mixtures. These have already been published, or are available in draft form from various professional organizations.

The DOE-YMP, has failed to recognize and account the scientific importance of complex mixtures and their health effects even though it has been brought to their attention. Failure to estimate possible health effects from complex mixtures can lead to very serious errors in assessing what is the real health risk associated with YMP. In sharp contrast, the France's nuclear waste management program has indicated that they are considering the health impact of complex mixtures in repository design as a serious health hazard. The DOE-YMP must recognize the scientific importance of complex mixtures and their potential health effect while considering the development of underground nuclear waste repository sites.

## V. RECOMMENDATIONS AND CONCLUSIONS

We recommend that the U.S. Department of Energy implement a research study of complex mixtures of radionuclides and hazardous chemicals using Physiologically-Based Pharmacokinetic Modeling (PBPK) and mechanistic approaches. This study might for example, measure the production levels of ROS by chemiluminescence bioassays (CL) after exposure of various mixed hazardous agents. The PBPK model takes into account species, intake route, metabolism, and excretion through a mathematical analysis describing these parameters for a variety of complex mixtures. PBPK models have been recommended by the National Council on Radiation Protection (24) and the International Commission on Radiological Protection (30). The use of PBPK modeling has also been recommended by EPA (16).

An upgrade of the TSPA model is needed to accommodate new emerging scientific data such as the "bystander effect": the term used to describe the biological effects observed in cells that are not themselves traversed by a charged particle, but are neighbors of cells that cause a genetic instability. This phenomena is observed at a dose as low irradiation of 1cGy (100 mRed) reported by Iyer and Lehner (31). The TSPA should also accommodate and the "adaptive response" reported by Sawant et al., (32). The potential health risk and risk assessment from bystander discussed Goldberg and Lehnert (33); and Oesterreicher et al., (34). Additionally, Mothersill and Syoumr (35) stated that: "over the past 15 years, it has only recently become apparent that chemicals in the natural environment also induce a state of genomic instability in cells and hence low dose chemical toxicity probably also involves bystander effects."

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