Project ID: **59925**

Project Title: **Modeling of Diffusion of Plutonium in Other Metals and of Gaseous Species in Plutonium-Based Systems**

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(ID 59925) Modeling of Diffusion of Plutonium in Other Metals and of Gaseous Species in Plutonium-Based Systems

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Research Objective:

The problem being addressed is to establish standards for temperature conditions under which plutonium, uranium, or neptunium from nuclear wastes permeates steel, with which it is in contact, by diffusion processes. The primary focus is on plutonium because of the greater difficulties created by the peculiarities of face-centered-cubic-stabilized (delta) plutonium (the form used in the technology generating the waste).

Temperature is the key controllable diffusion processes, i.e., temperature controls the rate of diffusion. The scientific goal of this project is to predict diffusion constants on an ab initio basis, i.e. diffusion distances in specified time at specified temperature for plutonium from plutonium-based waste materials into various steels or technologically-pertinent metallic alloys. This predictive ability will help to provide information relevant to setting temperature standards for maintaining structures, ducts, equipment or waste-containing vessels until such time as decontamination and decommissioning and/or permanent storage can be carried out. In addition, this knowledge will aid in assessing the depth of penetration that must be dealt with in any surface treatment for decontamination.

The scientific steps of the methodology are (1) to recognize the stabilizing mechanism and the electronic structure pertinent to that stabilization for face-centered-cubic (fcc) delta-stabilized plutonium, (2) to extract the information needed to perform dynamic simulations from ab initio electronic structure calculations, (3) to perform and report the dynamic simulations predicting the diffusion behavior.

Research Progress and Implications:

This report summarizes progress after 1 1/2 years of a 3-year project. We have completed step (1) recognizing the stabilizing mechanism and the electronic structure pertinent to that stabilization for face-centered-cubic (fcc) delta-stabilized plutonium. As part of this progress we have identified the roles and operating mechanisms both for temperature and for a stabilizing trivalent additive such as gallium. A publication appeared in May, 1999 reporting the way in which delta-stabilization occurs through a phase transition to a solid-solution-like phase involving a disordered mixture of two types of plutonium sites (para and ortho plutonium) having the same lattice symmetry but differing 5*f* electronic behavior. We have performed steps (2) and (3) for 3*d* transition metal alloys to develop the necessary methodology and to provide benchmark results for predictability of experimental behavior; and we have developed a detailed technical plan for proceeding with steps (2) and (3) for delta-stabilized plutonium in intimate contact with steel or other structural alloys.

We will be able to predict the migration distance of plutonium in steel in specified time at specified temperature. In addition, we will have the information and methodology needed to predict all thermoelastic properties of delta-stabilized plutonium and materials containing delta-stabilized plutonium.

There are two related uses for the results. The immediate use is to help in designing D&D efforts by predicting the effects of temperature and time on the location of plutonium in steel (or other structural material) building, equipment, or storage components. The intermediate-term use is in conjunction with developing and using a portable apparatus to "look through" steel using x-ray absorption spectroscopy to obtain detailed information about the detailed chemical and physical state of plutonium-contaminated material with a quantity and quality of detail well beyond that provided by any method and apparatus presently funded and scheduled for demonstration.

This solution to the problem of assessing and monitoring actinide contamination is necessary, and this approach is better than current technology, because plutonium is a very toxic radioactive material. Decontamination and decommissioning is a very difficult and costly process that must be done safely. The quality of information on the physical and chemical nature and distribution of the plutonium to be provided by the immediate computational modeling predictions and by the proposed portable in-situ characterization apparatus will make this process faster, less expensive, more reliable, and safer.

Planned Activities:

As stated above, we have developed a detailed technical plan for proceeding with steps (2, extracting the information needed to perform dynamic simulations from ab initio electronic structure calculations) and (3, performing and reporting the dynamic simulations predicting the diffusion behavior). We currently are proceeding systematically through the detailed parts of this plan.

We plan a three-phase approach to end-use. That end use will be to have one or more industrial companies use the plutonium (uranium and neptunium) characterization capabilities which we are developing, (and which we plan to demonstrate), in their D&D work at sites such as Savannah River, Hanford, and Rocky Flats. The first phase consists of the present modeling efforts and of reporting our results.

As the second, bridging, phase necessary to have methodology and apparatus to be employed by industrial companies in their work at the contaminated sites we need funding and an industrial partner (who will use our results) to join us in developing a portable XANES (x-ray absorption near edge spectroscopy) apparatus. We plan to upgrade and employ our rotating anode x-ray source. This will have sufficient x-ray intensity in the relevant spectral range to penetrate steel of the thickness used in ducts, building components, equipment, and containers.

To carry out the second phase of this plan, we need an industrial partner to join us in presenting DOE EM with a proposal/plan that will 1) develop the portable XANES apparatus, 2) demonstrate its utility at one of the sites, 3) make clear a plan for its use in the practical on-site D&D work. The end use will then be in the hands of the industrial contractor carrying out the D&D work.

This will enable us, in the third phase, to demonstrate and deliver for end use an operational technology to do two things. First is to assess the scope of a needed D&D effort. The on-site monitoring apparatus will provide a picture of the actinide species distribution in contaminated equipment and buildings, and our modeling capability will allow us to do time projections as to how that picture will change over a period of years under specified temperature conditions. For example, this might be the depth distribution of plutonium or other actinides below the surface of a component. Then the second use, would be to monitor the success in removal by laser ablation of the contaminated surface material. If desired, one could model a second time projection after the removal.

Information Access:

Publications:

"From Heavy Fermions to Random-Localized-Site Behavior Via Anderson Localization", to appear in Philosophical Magazine B 79, No. 5 pp 683-702 (May, 1999). (B.R. Cooper with O. Vogt, Q.G. Sheng, and Y.L. Lin).

"Calculated Lattice Relaxation in Pu-Ga Alloys", Journal of Alloys and Compounds 271-273, 367 (1998). (B.R. Cooper with J.D. Becker, J.M. Wills, and L. Cox).

"Calculated Lattice Relaxation in Pu-Ga", Phys. Rev. B 58B, 5143 (1998). (B.R. Cooper with J.D. Becker, J.M. Wills, and L. Cox).

"Correlation Effects on Stability in Pu Metal and Its Alloys", to appear in Electron Correlations and Materials Properties (A. Gonis and N. Kioussis, Eds., Plenum Publishing, 1999) (B.R. Cooper with P.E.A. Turchi, A. Gonis, N. Kioussis, and D.L. Price.

Invited Talks at International Conferences:

"Treating Electronic and Magnetic Properties of Actinide-Based Materials Beyond One-Electron Dynamics", School of Actinide Physics and Chemistry, Uppsala, Sweden, May, 1998.

"Synthesis of Many-Body Theory and Electronic Structure", International Workshop on Electron Correlations and Materials Properties, Heraklion, Crete, Greece, June 28-July 3, 1998.

Contributed Presentation at Conferences:

"Structural Relaxation in Pu-Ga via Full-Potential LMTO Calculations", Actinides 97 International Conference, Baden-Baden, Germany, September, 1997. (B.R. Cooper with J.D. Becker, J.M. Wills, and L. Cox).

"Diffusion of Plutonium into Transition Metallic Alloys and of Transition Metal Species into Plutonium", Eighth Conference on Computational Research on Materials, Lakeview, WV, May, 1998. (B.R. Cooper with S. Beiden).

"Modeling of Diffusion of Plutonium in Other Metals and of Gaseous Species in Plutonium-Based Systems", Environmental Management Workshop, Chicago, July, 1998. (B.R. Cooper with S. Beiden).

"Portable Detection and Analysis of Plutonium Content", Workshop on Environmental Management Science: Integration with End User Needs, Savannah River Site, November, 1998. (B.R. Cooper with D. Lederman).

"Modeling of Diffusion of Plutonium", Workshop on Environmental Management Science: Integration with End User Needs, Savannah River Site, November, 1998. (B.R. Cooper with S. Beiden).

"Modeling of Interdiffusion of Plutonium and Other Metals", Materials Research Society Meeting, Boston, November, 1998. (B.R. Cooper with S. Beiden).

"Anomalous Electronic Behavior and Relationship to Thermostructural Behavior of Light Actinides", American Physical Society Meeting, Atlanta, March, 1999.

"Electronic Structure of Alpha and Delta Plutonium", American Physical Society Meeting, Atlanta, March, 1999. (B.R. Cooper with N. Kioussis, P.E.A. Turchi, A. Gonis and D.L. Price).

"Equilibrium Lattice Volume of fcc Pu", American Physical Society Meeting, Atlanta, March, 1999. (B.R. Cooper with E.H. Sevilla and G.W. Fernando).

"Random 5*f* Localization and the fcc Transition and Depression of Melting Temperature in Plutonium, 29th Journees des Actinides Conference, Luso, Portugal, April 1999.