ALTERNATE MEASUREMENTS OF BENEFIT TO CRITICALITY ISSUES AT HANFORD

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ABSTRACT

Measurements in a critical mass facility, such as the Los Alamos Critical Experiments Facility, could be performed on simulated nuclear waste materials that would provide important critical mass information and concurrently provide a calibration of alternate measurement techniques. In addition to criticality information, the measurements could also provide a better assessment of diluent material/neutron interaction cross sections.

At Hanford, large quantities of fissionable materials are dispersed in tanks, fuel storage pools, and in solid waste. Although the fissionable materials are well diluted by a variety of neutron-moderating and -absorbing substances, it is difficult to assess the margin of subcriticality. A number of measurement approaches are proposed that will either help determine fissionable material concentrations, distributions, or provide a direct measure of subcriticality. The methods under consideration involve passive neutron counting, active neutron measurements, pulse neutron applications, neutron noise analyses, and cover gas evaluations. Active neutron measurements can also provide insight into the determination of neutron absorber concentrations. Efforts are underway to test some of the methods in actual waste tank environments and geometries. It is important that these methods be tested and calibrated in a critical mass facility.

The information derived from these critical mass measurements would not only be of value to the Hanford Site but throughout the industry where criticality concerns are related to safe handling of waste materials. The "Forecast of Criticality Experiments and Experimental Programs Needed to Support Nuclear Operations in the United States of America: 1994-1999" identifies a series of experiments – 502, 502a through 502i – that describe needed criticality measurements associated with nuclear waste handling.

BACKGROUND

The formal implementation plan for recommendation 93-2 of the Defense Nuclear Facilities Safety Board recognizes that a special-purpose experiments program operating within a general-purpose criticality facility is an ingredient of an effective criticality safety program. For environmental restoration and waste management activities in which criticality safety issues come into play, the question is raised: what exactly is the relationship between such a facility and those issues? This is a valid question that ought to be carefully addressed if effective use is to be made of a general-purpose criticality facility. Clearly, the answer depends on (a) what precisely is the nature and extent of the criticality issues involved and (b) how exactly does a critical facility contribute to the resolution of these issues? Assuming that answers to those questions have been provided, an adjunct to those questions might well be: are there situations in which a criticality facility can be used in an optimal way so that the issues in question can be resolved in the most effective manner? One situation in which this last question comes into play is the use of the Los Alamos Critical Experiments Facility (LACEF). Use of the LACEF might be optimized if it could be applied in conjunction with alternative measurement approaches at Hanford and other DOE sites in a way that the resolution of criticality issues at those sites would benefit the most.

Large quantities of nuclear waste at different U.S. Department of Energy (DOE) production sites contain fissionable materials. In many instances the concentration and distribution of fissionable material in the waste, as well as the chemical composition, are not well known. The lack of such information makes it difficult to implement nuclear criticality safety controls. Chemical and isotopic analyses of waste are a possible, but a very costly, option. More expeditious approaches to waste nuclear criticality control could be based on *in situ* measurements estimating fissionable material and neutron-absorber concentrations. Such measurements need to be performed with instrumentation that can be calibrated in simulated materials in environments and mockups that lend themselves to computer modeling. To establish margins of subcriticality, parametric measurements are needed in simulant materials that look at neutron multiplication values as a function of fissionable material or nuclear diluent and absorber concentrations. A facility, such as a critical mass laboratory, is required for these measurements.

The *in situ* measurements are of specific importance to Hanford. Two major unreviewed safety questions (USQ) – one in the tank farms and the other in the KE Basin – have been related to nuclear criticality limits compliance issues. Although the tank farm criticality USQ has already been resolved, with applicable calibrated measurement techniques in place, the resolution of these USQs could have been expedited or prevented the USQ declaration.

Over the 42 years of nuclear materials production, significant quantities of plutonium and uranium ended up in waste streams. The 177 waste tanks at Hanford are estimated to contain as much as 1,000 kg of plutonium and significantly more uranium. In addition to the waste tanks, plutonium can be found in cribs, solid waste, spent fuel, and sludges in spent fuel pools. The concentration of the plutonium and the geometric configuration, moderation, and control by neutron absorbers is not well known.

Although the waste tanks were shown to be safe in the current configuration, based on analyses, future processing of tank wastes will perturb existing conditions. Monitoring and surveillance instrumentation need to be in place to demonstrate that criticality margins are not compromised. Because the tank waste subcriticality depends in part on neutron absorbing diluents, their presence, along with the impact of diluents on critical masses, needs to be known. Measurements of subcritical and critical configurations with substantial amounts of SiO_2, MgO, CaO_2, and nitrogen compounds would be highly desirable. Correlation of the measurements with computer codes would resolve cross-section ambiguities. Alternate measurement techniques, such as the application of pulse neutron systems, could provide very useful insights into diluent and absorber concentrations, in addition to fissionable material concentrations.

MEASUREMENT APPROACHES

There are very limited critical mass data for plutonium and uranium as chemical compounds and as part of a waste matrix. Critical mass measurements are needed for prototypic waste material and a geometry, both of which can be readily replicated with computational tools and cross-section sets. As critical mass measurements are performed, alternate techniques for monitoring subcritical conditions could be calibrated. Eventually, such alternate techniques will provide *in situ* surveillance for compliance to nuclear criticality limits. The experiments should initially consider simple geometries, such as cylindrical tanks filled with homogeneous materials. The tanks would have limited access for measurement devices both on the outside and internal to the tank. Support facilities should have the capability to build up gradually fissionable material concentrations within the tank and add in nuclear diluents and absorbers.

Fissionable materials can be detected as a result of the fission process and transmutations. Characteristic gamma rays, alpha particles, neutrons, and gaseous fission products that are given off by the nuclear processes can serve as detection indicators. If the gamma-ray background radiation is strong, such as in a typical waste tank (10 - 600 rad/h), neutron-based methods should be more viable.

Because of the high gamma-ray background, the concepts considered are mainly neutron based. Most are differential measurements in that they will tend to characterize materials in a narrow cylinder around the observation well or interrogation port. They will, however, provide very useful data on distribution gradients in the axial direction. The concepts considered involve passive neutron counting devices, such as track recorders and activation foils; active systems using neutron sources and detectors; and systems consisting of a pulsed neutron generator and detectors.

In addition to the differential measurements, integral measurements are being considered that quantify certain gaseous fission products being generated in the waste tanks. Gases such as helium, krypton, and xenon would be characteristic of total transuranic or fissionable material content. Certain other gases, such as radon, could provide information on uranium contents.

Fission track recorders consisting of highly enriched uranium foils imbedded between sheets of mica would record fission product tracks following neutron-induced fissions in the foils. The tracks can be counted and related to a neutron density. Such methods were successfully used in the Three Mile Island core recovery efforts. The advantages of fission track recorders are that they require no special electronics, can be readily deployed, and can remain in place over extended time periods. The tracks in the mica become a permanent record. The disadvantages are that neutrons are detected that arise either from (α,n) reactions or fissions and that special computerized optical equipment is needed to read the mica sheets.

Metallic foils such as copper, indium, zinc, and magnesium are very similar to fission track recorders except they may have short saturation times and, again, require some highly specialized equipment for counting. With the right combination of foils, it may be possible to discriminate neutrons coming from fissions versus (α,n) reactions.

Use of neutron counters such as BF_3 or ^3He can be employed for passive neutron detection. The advantage of the detectors is they can provide instantaneous readout and can be readily repositioned to obtain scans of neutron distributions. With filters on the tubes, some discrimination as to neutron origin is possible.

Active neutron sources such as ^{252}Cf or pulse neutron generators can be employed to measure low concentrations of fissionable materials and also provide some assessments of the subcriticality of test configuration. ^{252}Cf-source-driven neutron noise analyses techniques have been employed by J. T. Mihalczo to measure neutron multiplication factors as low as 0.3. Active methods can resolve the origin of neutrons and even provide insights into the concentrations of other materials, such as moderators or neutron-absorbing diluents in the matrix. Disadvantages of such systems are the cost, the size of equipment, and the fact that measurements cover localized regions.

Another measurement concept that could provide useful information is gas analyses for helium and fission products. Alpha-particle decay in nuclear waste produces helium gas, while spontaneous and induced fissions create noble gas fission products. The gases produced by fissionable material in a matrix will tend to migrate through the material and collect in air spaces. Sampling of the air for these special constituents will tell if the system is generating gases in excess of natural background. An excess can be correlated to a fissionable material content. A gas measurement would represent an integral fissionable material content of the system. High-precision mass spectrometers, as well as radioactive material detectors, are needed for measurements. Some disadvantages of this method are the requirement for very sophisticated gas analyses and control over the airflow in the air space.

Neutron activation and detection of characteristic gamma rays is a viable measurement alternative, provided a background gamma-ray field does not exist. In the Hanford Site waste tanks, dose rates of hundreds to 1,000 rad/h are encountered, therefore, gamma-ray-based techniques may be less successful.

EXPERIMENTAL MEASUREMENTS

The objective of alternate measurement techniques is to detect very low concentrations and distributions of fissionable materials in nuclear waste. A typical range would be 0.05 g/L to 3 g/L. The range is below subcritical measurement capabilities. However, if the concentration in tank waste at these low levels can be determined, an assessment of the total fissionable inventory in the tank can be made. Current plans call for intercomparison of measurement techniques in the tanks supplemented with some criticality computations and chemical sample analyses. What is needed is a well-characterized system in a controlled environment of a critical mass facility such as the LACEF. An experiment in a tank containing a simulated waste matrix is envisioned where the concentration of fissionable material is gradually built up. At each concentration change, measurements with passive and active neutron systems are made. As the system neutron multiplication increases, gas analysis would be attempted. Noise analysis (pulse neutron) would track the approach to critical and provide a much needed intercomparison between methods. Perturbations on the basic experiment could include diluent material changes and subsequent approach to critical. In such a measurement, multiple important objectives could be met to help provide much needed benchmark and calibration data to nuclear waste criticality problems.

More complicated systems are envisioned for subsequent measurements that would require mockups of distribution gradients, fissile material layering, positive reactivity insertions due to mixing, and accounting for localized concentrations of fissile materials. The experimental data, calibration of methods, and calculations based on such information should help in demonstrating compliance with DOE Order 5480.24, especially in the area of risk determination.

CONCLUSIONS

It is important that a nuclear criticality laboratory be available to provide measurements for nuclear waste, a multicomponent chemical system about which little is known. Simultaneous with the critical mass measurement during the approach to critical, a number of *in situ* measurement techniques can be calibrated and intercompared. Perturbations on the waste composition could also furnish insights into cross-section uncertainties of diluents in waste materials and their impact on the neutron multiplication constant. Significant benefit to a broad spectrum of sites could be derived from such an experimental program in terms of enhanced nuclear criticality safety implementation and possible cost avoidance.