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July 16, 2001

Mr. Carl Will
State of New Mexico Environment Department
Hazardous Waste Bureau
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Reference: Work Assignment No. Y513, 06082.600; State of New Mexico Environment Department, Santa Fe, New Mexico; General Permit Support Contracts; NMED-HSW Corrective Action Chapter of the RCRA Permit Renewal for the Los Alamos National Laboratory; Research of monitoring, remediation and dose assessment techniques for Material Disposal Area W (MDA-W), Technical Area 35 (TA-35) at Los Alamos National Laboratory (LANL); Task 8 Deliverable

Dear Mr. Will:

Enclosed please find the deliverable for the above-referenced work assignment. This deliverable consists of a discussion of MDA-W, which consists of the two sodium potassium coolant-filled tubes suspended within a carbon-steel lined casing set down into the Bandelier Tuff. The cooling fluid is contaminated with plutonium and fission products. The deliverable discusses possible methods for monitoring the tubes for leaks, remediation techniques and dose assessment. The draft of the deliverable was e-mailed to you on Monday, July 16, 2001, at carl_will@nmenv.state.nm.us. The deliverable is formatted in Microsoft Word 2000.

If you have any questions, please feel free to contact Ms. Paige Walton at (801) 582-9329.

Sincerely,

June K. Dreith

June K. Dreith
Project Manager

32383

Enclosure

cc: Mr. John Young, NMED
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# TASK 8 DELIVERABLE

# RESEARCH OF MONITORING, REMEDIATION AND DOSE ASSESSMENT TECHNIQUES FOR MATERIAL DISPOSAL AREA W (MDA-W), TECHNICAL AREA 35 (TA-35) AT LOS ALAMOS NATIONAL LABORATORY (LANL)

**NMED-HSW Corrective Action Chapter of the RCRA Permit Renewal for the Los Alamos National Laboratory**

**Submitted by:**

**TechLaw, Inc.**
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**Submitted to:**

**Mr. Carl Will**
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**Santa Fe, New Mexico 87505**

**In response to:**

**Work Assignment No. Y513, 06082.600**

**July 2001**

## 3.0 NATURE OF CONTAMINATION

In order to determine the best possible options for monitoring the tubes for potential leaks and assessing the overall condition of the tubes, some assumptions must be made concerning the sodium cooling fluid. Information from the SWMU Report (LANL 1990) indicates that tubes contain 500-650 pounds of a sodium potassium (NaK) alloy of unknown composition, as well as plutonium (Pu-239, Pu-240 and Pu-238) and fission products (Cs-137, Ni-59, Co-60 and Na-22).

Pure sodium (Na) has a melting point of approximately 98°C (208.4 °F) and potassium (K) has a melting point of about 63°C (145.4°F). This means that if the tubes were to contain pure sodium, the material within the tubes would be a solid at ambient temperatures. However, the tubes are reported to contain an alloy of sodium and potassium (NaK). The melting point for NaK will vary depending upon the fraction of Na and K in the alloy. The most commonly used NaK alloy is the lowest melting point composition (22% Na and 78% K), which has a melting point of 12 F. At or near room temperatures alloys near this composition will be liquids. Neither the exact composition of the metal nor its physical state (liquid or solid) is known. It is assumed (for conservatism) that the mixture within the MDA-W tubes is a liquid, but it is also possible (but we think less likely) that it is a solid. Monitoring and remediation options (discussed in Sections 4 and 5) will discuss both liquid and solid states.

Another unknown about the nature of the coolant fluid is the concentrations of plutonium and fission products. One of the LANL references (LANL 1992) indicates that the plutonium concentration is low, at less than 0.1 parts per million (ppm). However, none of the available references indicated concentrations of the fission products. While in the long term plutonium is a concern, especially for internal exposures, for external radiation exposure hazards, the concentrations of the fission products are of greater concern. This is because plutonium is an alpha emitter and the alpha radiation cannot penetrate the stainless steel tubes. Thus, if the tubes are intact, there is little concern over external radiation exposure due to the plutonium. On the other hand, the fission products emit beta and gamma radiation (Cs-137, Na-22 and Co-60) or gamma radiation (Ni-59). The activity of the coolant and amounts of fission products are unknown. The stainless steel tubes will shield against most of the beta radiations, but for the most part, the steel will not significantly attenuate the gamma radiation. Therefore, there is concern from external radiation exposure due to the fission products. The greater the concentration of the fission products within the tubes, the greater the potential exposure rate.

If there is a leak of the coolant from the tubes, the potassium in the NaK can react with any atmospheric oxygen in the annulus to form three oxides: potassium oxide, potassium peroxide and potassium superoxide. These oxides would form a crust over the exposed NaK surface. If a disturbance (i.e., movement, water, etc.) would break the seal of the crust over the NaK, and the potassium superoxide mixes with the potassium in the NaK fluid, a high-temperature thermite reaction can occur. This reaction can be enhanced and become quite explosive by the addition of a fuel, such a petroleum product or drilling fluid. The speed of the reaction is dependent upon the atmospheric conditions within the annulus. Since the tubes and casing are below ground and sealed at the top with concrete, it is not likely that there is much oxygen from the air trapped within the annulus to perpetuate the formation of a significant amount of superoxides. However, this is an unknown.

Another reaction of concern if there has been a leak of the coolant is the potential for the formation of hydrogen gas. Sodium reacts with water to form a sodium hydroxide (highly alkaline caustic soda) and hydrogen gas. Buildup of hydrogen gas could be problem in the tubes, if water has been introduced into the system. Again, this is an unknown and potential safety hazard.

## 4.0 MONITORING

The question of how to monitor the MDA-W tubes raises several concerns. The first concern is how to keep the integrity of the steel tubes and the casing intact. The second is how to monitor the system without creating a conduit for water to enter the system. A third concern is how to monitor the system given the unknowns about the physical state of the coolant and amount of radiation. A fourth concern, or rather a challenge, is accounting for potential hazards to personnel monitoring the system by incorporating necessary risk precautions for exposure to radiation, any formed potassium superoxide, and hydrogen gas.

### 4.1 Monitoring Techniques for No Further Consideration

One alternative for monitoring the system is to employ angled directional drilling to bore a small conduit down to the base of the open annulus. From this borehole, samples could be taken to determine if a leak has occurred in the past, and periodic samples could also be taken to monitoring for leaks. However, by drilling a borehole, a direct conduit for water and air (oxygen) would be created. It seems highly unlikely that it would be possible to completely seal this boring from any water entering the system. Therefore, this idea of a directional boring to the bottom of the annulus will not be considered further.

Another alternative is to drill a small hole thought the concrete cap and into the annulus, and install a small valve at the top of the hole. From this valve, periodic air samples could be taken and evaluated for levels of hydrogen gas. Sodium when exposed to water will form a sodium hydroxide and hydrogen gas. If the tubes have leaked sodium coolant, the sodium would react with any water present in the system forming hydrogen gas. If increases in hydrogen gas were noticed in monitoring, it could be assumed there was a leak in one of the tubes. However, this system does not appear practical, as evidenced from the LANL hydrology studies all indicate that groundwater in this area is not a concern, and that the tubes are most likely in a dry environment so detection of a leak cannot be assured.

Radiation monitoring on the surface would probably not be sufficient to determine if a leak from the tubes has occurred. While it is noted that the LANL RFI report indicates that the concrete has a surface reading of 10 uR/hr, concrete also has a natural background radiation associated with it due to the presence of NORM (naturally occurring radioactive materials) in the materials that comprise concrete. Without a background reading from similar concrete, the radiological data as presented in the LANL document has no meaning. The primary radiological constituents in the sodium cooling fluid are plutonium (pure alpha emitter), cesium-137 (strong energy beta, low to medium energy gamma emitter), sodium-22 (medium energy beta and gamma emitter), cobalt-60 (medium energy beta and gamma emitter) and nickel-59 (low energy gamma emitter). The alpha radiation from the plutonium would be shielded by the steel tubes and casing, as well as by the tuff and concrete. There is not an effective way to monitor on the ground surface for the plutonium emissions. As for the beta and gamma emissions from the fission products, if the tubes did sustain a leak, it is likely that the cooling fluid would flow down the annulus. The concrete, steel casing and tuff would also successfully shield and attenuate most of the beta and gamma emissions. Therefore, it does not appear that surface monitoring using radiation detectors would be successful for monitoring to determine if the tubes have leaked.

4.2 Monitoring Alternatives for Further Consideration

There are two monitoring techniques when used in combination that seem to be both effective and low in risk: a combination of radiological monitoring within the annulus and use of a down-hole video camera. A small borehole would be drilled through the concrete and into the annulus. Some magnetic monitoring may need to be applied prior to drilling to determine the exact locations of the two tubes, so as not drill into them. The drilling would have to be dry-drilled. If the tubes have leaked and potassium superoxides have formed, the use of drilling fluids could act as a catalyst for a reaction. Once the hole has been drilled, it should be lined with a inert liner and capped.

A small down-hole video camera, as used for pipe inspections, would be lowered down the monitoring hole and into the annulus. Using the remote camera system would allow for periodic visual inspections of the tubes. This would also determine if there has been any leaks to date from the pipes, allow for the assessment of the integrity of the tubes and the carbon-steel liner and determine with time if any leaks occur. The video system would also allow for comparison of the tube conditions over time.

The video system would be coupled with radiation monitoring. The radiation detector to be employed should be a beta detector. A beta detector was chosen for the following reasons:

- Alpha radiation from the plutonium is too easily attenuated and shielded and detection based upon alpha radiation may not be reliable,
- Some of the gamma radiation may be able to penetrate the stainless steel tubes. Therefore, it would be difficult to discern the amount of detectable energy from within the pipe and from any potentially leaked material, and

- While some of the beta energy from the fission product may not be 100% attenuated by the stainless steel tubes, most of the beta emission would be attenuated, allowing for a low background reading. Any leak of material from the pipe would immediately show a jump in the amount of beta emissions, making a leak fairly easy to identify.

A background reading for the beta would need to be established. Determination of background may create the need to establish a similar type borehole lined with carbon-steel to simulate what natural background readings may be down-hole. However, based upon the visual inspection of the pipe using the video camera, it is determined that no leaks have occurred, a background may be established within the MDA-W system. Any noticeable variations in beta readings from the baseline would indicate a potential leak of cooling fluid. Background for this situation would include any beta that penetrates the tubing. Since it may be very difficult to establish a valid background from another hole where the tubing is not present, the camera survey that establishes there have been no leaks could validate initial readings as background.

Two other systems that should be given some additional consideration are the use of a bubbler tube and thermal switch. A bubbler tube would measures pressure differences to determine if liquid was present in the bottom of the tubes. The bubbler tube would be fed through the hole drilled in the concrete, as described for the radiation monitoring and video camera. A low flow of nitrogen or another inert gas is introduced into a dip pipe extending to near the bottom of the borehole. A pressure monitor measures the amount of backpressure on the nitrogen line. If the monitor does not measure any backpressure, then there is no fluid at the bottom of the tubes, and thus no leaks. The amount of backpressure measured would directly relate to amount of fluid pooled at the bottom of MDA-W annulus. It would be necessary to vent the injected gas from the system through some kind of seal.

The other system, which could be applicable, is the use of thermal switches. Thermal switches would sense either the difference between temperatures in the void space of the annulus and any liquid or could read the difference in thermal conductivity as the probe becomes submerged in the liquid.

The bubbler tube and thermal switch would only be appropriate if a significant amount of cooling fluid had leaked from the tubes and had pooled within the annulus. This would assume that the tuff has relatively impermeable properties, which seem to be substantiated in the LANL geologic reports reviewed.

If the coolant had solidified, then there are no real immediate problems. The biggest concern would be the introduction of water and oxygen into the tubes. If the tubes have ruptured, the water could migrate into the tubes and react with the solid NaK forming hydrogen gas and caustic soda. Oxygen could cause a reaction with any potassium superoxide that may have formed. All of the monitoring techniques discuss above would apply if the coolant has solidified.

## 5.0 REMEDIAL OPTIONS

The remedial options depend on whether the sodium cooling fluid is a fluid or whether the mixture has solidified, and on the concentration of the fission products, how "hot" the material is.

If the material within the tubes has solidified, the tubes could be removed and shipped to an off-site storage facility. Which facility would depend on the concentrations of the fission products within each tube. If the material could be classified as a Class A waste, then the tubes could possibly be sent to Envirocare of Utah. However, if the material within the tubes is classified as a Class B/C waste, then the tubes would need to go to either Barnwell, South Carolina or the Department of Energy's Hanford Reservation in Washington. Great care would need to be taken in removing the concrete from around the tubes.

If the material within the tubes is a liquid, the tubes could be removed. Given the nature of a potentially mixed waste liquid, the only disposal facility option would be Yucca Mountain, which has yet to open. It is also not certain if Yucca Mountain will accept liquid wastes. It appears that the most reasonable remedial option for the tubes if the sodium cooling fluid is a liquid is to leave them in place and institute site controls and monitoring requirements.

A concern about leaving the tubes in place is the location of MDA-W in relation to the edge of the Ten Site Canyon. Based upon the information in the available reference documents, it could not be determined the exact location of MDA-W in reference to the canyon edge/walls. This concern is raised as several documents reviewed for TA-54 indicated that mass wasting of the canyon walls is a large concern. It appears that MDA-W is located near the canyon wall of Ten Site Canyon. Do the concerns of mass wasting and erosion of the canyons around TA-54 also apply to the Ten Site Canyon at TA-35?

Research is being conducted at the Oak Ridge National Laboratory Y-12 facility on separation of radionuclides from aqueous wastes. Some studies are currently being conducted on how to separate dissolved radionuclides (primarily fission products) from liquid solutions (supernatants) with high concentrations of sodium and potassium salts using various systems of filters, ion-exchange materials and sorbents. The work is currently on going, but could result in a viable technique that could be applied to the cooling fluids in the MDA-W. If technology becomes available, it may be possible to separate the radionuclides from the cooling fluid, thus increasing disposal options.

## 6.0 DOSE ASSESSMENT

Two of the primary reasons for assessing and recording radiation exposures are to ensure that workers/residents (receptors) are receiving adequate protection and demonstrating compliance with regulatory limits. There are two pathways of exposure that result in doses and associated risks: internal and external exposure. Potential exposure to the plutonium and fission products in the cooling fluid would occur through these pathways only if there were a leak in the tubes, allowing for migration of radionuclides to or near the ground surface (i.e, the hole in the concrete could create a conduit for both internal and external radiation exposure), or by excavation of the tubes for disposal (i.e, intact tubes would result in external exposure only). If the tubes are intact and no leakage of the coolant has occurred, then risks from inhalation exposure are non-existent. The tuff and outer steel casing provide adequate shielding to mitigate any external exposures from material within intact tubes.

Internal or inhalation radiation dose differs from an external dose in that internal radiation doses result from the actual intake and deposition of radioactive material in the body. This material may be retained in the body, depending on the half-life of the radionuclides, and may result in doses over many years after the initial intake, due to the retention of the radionuclides within the body. External radiation doses occur when a person is near a radiation source, and the body absorbs the radiation emitted from a radioactive source. Once the person moves away from the source or utilizes shielding, the exposure is mitigated. However, both internal and external exposures are equally a concern to the safety of potential receptors.

There are radiation dose models that can be used to calculate doses from exposure to radionuclides. For example, the model RESRAD, developed through Argonne National Laboratory, can be used to simulate the dose received to an individual from exposure in a residential or agricultural setting. RESRAD will determine both internal and external doses. However, it appears that the doses received from exposure to the radionuclides within the tubes can be determined through simpler means.

Inhalation doses would be most easily determined through simple spreadsheet calculations. Simplified, inhalation dose is a function of the concentration of the radionuclides, exposure duration and inhalation rate. For inhalation doses, the exposure to plutonium and the fission products is important, and the activity of all the radionuclides in the coolant would need to be determined prior to determination of potential doses. Internal doses can also be monitored by bioassay techniques.

For external radiation exposure, either spreadsheet calculations or the Grove Engineering model Microshield could be applied. The external exposure would come solely from the beta and gamma emitting fission products, as the alpha emitting plutonium would result in a negligible external dose. Both the model and a simple spreadsheet calculation would determine the external dose as a function of the amount of radiation, the size or area of contamination, the distance from the source the person is, the exposure time and any potential shielding between the person and the source. Since it is not known the concentration or activity of the fission products contained in the tubes, the external dose could not be determined at this time. External doses may also be monitored through the use of thermoluminescent dosimeter (TLD) badges. The TLD is worn on the outside clothing of a person to record the amount of external radiation exposure. Typically TLD badges are sent monthly into a laboratory for analysis.

## 7.0 REFERENCES

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