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INVESTIGATION OF CASK CONTAMINATION WEEPING A PROGRESS REPORT*

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Investigation of Cask Contamination Weeping:
A Progress Report

1.0 INTRODUCTION

Spent fuel transportation casks have arrived at final destinations with removable surface contamination levels in excess of regulatory limits [1,2], although pre-transport surveys indicated removable contamination levels were well below these limits. The control of this in-transit "weeping" of surface contamination on pool-loaded spent fuel transport casks is of particular concern to both the U.S. Department of Energy (DOE) and the U.S. Nuclear Regulatory Commission (NRC).

Weeping, also known as sweating, is the transformation of fixed radioactive particulates on an exterior surface of a transport cask to a removable state. Weeping has been observed sometime after a cask is removed from a fuel pool and decontaminated. The weeping phenomenon is countered by time-consuming operational constraints and procedures which have a significant impact on cask turnaround times and occupational exposures at transport facilities. Further, the arrival of a contaminated cask results in negative public perceptions that are inconsistent with DOE and NRC goals.

The objectives in resolving the technical issue of weeping are to identify specific causes of the weeping phenomenon, then to implement new cask design requirements and supporting operational procedures which will limit or inhibit the accumulation, retention, and in-transit conversion of fixed surface contamination. Benefits of finding a solution to weeping therefore include:

- Reduced contamination of casks
- Improved decontamination effectiveness
- Reduced occupational exposure
- Reduced cask turn-around times.

Such objectives will require an understanding of the physical processes and the determination of related parameters which contribute to the weeping phenomenon. This paper describes current efforts at Sandia National Laboratories (SNL), under the auspices of the DOE Office of Civilian Radioactive Waste Management, to resolve the contamination weeping issue.

2.0 APPROACH

The approach to the problem consists of three elements: 1) the collection of available data; 2) theoretical development of a physical model; and 3) experimental evaluation of the model and available data.

2.1 Field Data

The first element is the compilation and evaluation of existing tranport data, to determine the extent of the problem and, where possible, which parameters contribute to the initiation, rate, and continuance of the weeping phenomenon.

Compilation of transport data has been carried out by Transnuclear, Inc. (TN), Chem-Nuclear Systems, Inc. (CNSI), and SNL.

2.2 Physical Model Development

The second element to the approach is a theoretical treatment of parameters suggested by the transport data to influence the weeping phenomenon. Chemical bonding and transport effects may be influenced by contaminant type, concentration, and chemical form. Environmental conditions such as temperature, humidity, contaminant exposure time, and cask surface condition may also contribute to the weeping phenomenon. The University of Texas at Austin is performing the physical model development.

2.3 Experimental Evaluation

The third element in the approach is an experimental evaluation of the physical model, with some parameters suggested by the transport data. Experiments will consist of inducing weeping under controlled conditions: varying suspected environmental and material parameters and then correlating the parameter variations with observed removable contamination. Exposure to contaminants will be accomplished by submerging test cylinders of various materials and finishes in the Union Electric Callaway Plant power reactor spent-fuel storage pool. Weeping will be measured by documenting the rise in removable contamination periodically throughout the procedure.

3.0 RESULTS AND DISCUSSION

The contamination weeping investigation is currently underway; although data collection and analysis are not yet complete. Selected data together with a preliminary analysis are presented below. A brief description of the theoretical model is also presented, as is a description of the experimental procedure being used.

3.1 Data Collection and Analysis

Transportation records were searched for weeping incidents in the U.S., and current transportation campaigns have been monitored to determine factors involved in the occurrence of weeping.

3.1.1 Background

The Radioactive Materials Incident Reporting (RMIR) database contains publicly available information about radioactive materials transportation incidents since 1971. These were drawn from the Hazardous Materials Incident Reporting System of the U.S. Department of Transportation (DOT), the NRC files, and from state radiological control offices. From January 1977 through July 1986, the RMIR reports 46 cases of excessive removable surface contamination. These were all pool-loaded Type B casks, loaded or empty, carrying spent fuel or irradiated hardware, and exhibiting over 20,000 dpm/100 cm^2 removable contamination.

Experience with recurring weeping episodes has lead the NRC to term the phenomenon "chronic"[3]. From 1983-1987, NRC inspectors, routinely engaged in inspection of spent fuel tranports, observed a recurring problem of excessive surface contamination at cask destinations. Several dozen cases of contamination weeping were reported in which removable contamination was found on arrival to exceed regulatory limits, even though these limits were not exceeded prior to tranport.

3.1.2 Campaign observations

The most complete set of spent fuel transportation campaign data to date is that provided by Transnuclear, Inc. (TN). From December 1985 to June 1986, TN carried out two campaigns in which spent BWR fuel was moved from the West Valley Demonstration Project to the Dresden and Oyster Creek power stations. Dates, routings, and contamination levels were available from these campaigns. Weather information was also reported, and a weather profile reconstructed for each tranport.

Regression analysis of these data indicated a model of the following form:

$$\text{Log } C = \mu + A + \beta_1 H + \beta_2 D + \varepsilon,$$

where:
- C is the removable contamination level
- μ is the mean contamination rise
- A is a factor which is dependent upon the combination of the transporting and receiving facilities
- H is the daily high temperature
- D is the daily temperature differential (high - low)
- β_1 and β_2 are weighting factors
- ε is the error associated with the analysis.

The site-dependent term A appears to dominate contamination levels in this model. Factors which affect the value of A may include pool cleanliness and time of submergence, as well as differences in decontamination techniques, measurement techniques, and instrumentation. The high temperature term appears to be a strong factor, indicating that lower values of H accompany increased removable contamination. Similarly, the lower temperature differential values show a distinct, though weaker, correlation. Humidity and precipitation did not appear to have a strong effect on increasing removable contamination levels and are not represented in the final model.

3.1.3 Weeping on a cask in storage

Casks appear to undergo weeping not just in transit, but also in storage. Two Transnuclear casks, TN 8L.3 and TN 8L.4, have been under observation for weeping while in storage. Throughout the observation period, the casks were parked on their transporters and covered with canvas personnel barriers. Precisely marked positions were monitored on a bi-weekly basis by the same technician, and no decontamination was performed during the observations.

The TN 8L.4 cask was admitted on November 5, 1987, to a storage yard in Aiken, South Carolina, with removable contamination levels lower than 500 dpm/100 cm^2. On November 13, an area of the 304 stainless steel front drum exhibited removable levels in excess of 12,000 dpm/100 cm^2, which began a pattern of contamination oscillation shown in Figure 1. Swipes taken of the weeping surface at its peak yielded 85% Cs 137, 14% Ag 110m, and 1% Co 60.

The TN 8L.3 cask, which completed a transfer campaign between Duke Power Company's Oconee and McGuire stations on February 19, 1988, was admitted to the Aiken storage yard with removable contamination levels below 1000 dpm/100 cm^2. By the end of March, the level of one area on the front drum rose to 10500, as shown in Figure 2 (note: dpm/100cm^2 has been dropped for clarity of the discussion). It was then decontaminated and used in a campaign from April 14 to May 19, 1988. The levels after check-in at Aiken remained less than 1000 until August 1988 when they rose again to 9000. Levels then oscillated from near 3000 to 11000 (November 1988). Subsequent measurements were: December-- 5000; January-- 3900; February-- 3800; March-- 2800; April-- 5000.

To determine possible correlations between cask surface structure and weeping behavior, replications of the weeping and non-weeping TN 8L.3 surfaces were made for examination. The surfaces were covered with solvent-softened acetate films, which formed imprints of the surfaces. The films were then removed, sputtered with a Au-Ir target, and examined under a Scanning Electron Microscope (SEM).

Surface replications were made at three different times: July, October, and December 1988. Prior to the first replication, the weeping area was cleaned and decontaminated. The replication itself (RSJ) was damaged and unfit for SEM examination. The October replications (RSI series) consisted of two weeping and two non-weeping samples, taken at the locations indicated in Figure 2. Figure 3 shows the non-weeping surface. The striations could be a randomly scratched metal surface with distributed low-atomic weight debris. No distinct grain boundaries are visible in this surface. Figure 4 illustrates a relatively clear etched grain structure, which is part of a weeping surface.

The third set of replications of the same surfaces (RSII series) was made in December. The locations, also shown in Figure 2, correspond to Figures 5 and 6. The replicated weeping surface, represented by Figure 5, displays large striations and what appears to be fine scratches, as well as embedded particles. None of the replicas displayed the distinct grain structure seen in the previous set. The non-weeping replicated surface, shown in Figure 6, displays the same type of striated surface and debris as those of the weeping surface. The debris consisted primarily of stainless steel constituents Fe and Cr. Other debris included Na, Al, Si, Cl, K and Ca. Contaminants found by spectral analysis are listed in Table I.

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TABLE I. Surface Replication Contaminants

Sample	Contaminant (pCi) Cs 137	Cs 134	Co 60	K 40	Ag 110m	Total
RSI-J	479	86	31	7	17	620
RSI-C1	944	187	136		361	1628
RSI-C2	381	65	66	62		574
RSI-S1	2588	606	650	397	227	4468
RSI-S2	2132	427	1160	316	605	4640
RSII-C1	16			7		25
RSII-C2	2					2
RSII-S3	9	2				11
RSII-S4	13	3				16
RSII-S5	5			9		14
RSII-S6	20	3	1	9		33

The concentration of contaminants is greater in the RSI (October) replicates, just prior to the peak weeping level in November.

3.2 Theoretical Model

The theoretical model of contamination under development considers the formation of a surface passivation layer on materials typically used for cask outer skins. Passivation occurs when a stable oxide layer is formed on the surface of a metal such as stainless steel. Surfaces which are anodic due to electrode potential differences, stress and concentration cells, etc., will corrode in the presence of an electrolyte. This layer tends to isolate anodic surfaces and reduce anodic current density, and thus reduce corrosion rates to insignificant levels.

The formation and subsequent breakdown and reformation of the passivation layer may lead to inclusion and release of contaminants in the layer structure. Factors which may affect this process include cask material constituents, environmental conditions, and environmental constituents. For example, chromium in stainless steel will produce passivation because of its strong attraction for oxygen. An electrolyte is provided by humidity in the air or from pool water and allows corrosive action to take place. Moisture may also provide the vehicle by which contamination could pass into the layer through cracks, which could be formed by stresses due to thermal gradients, handling and cyclic loading during transport. The type and concentration of contaminants available for inclusion, as well as the presence of active ions such as chloride may affect the growth and composition of the film. The importance of each of these factors will be examined in the experimental phase of the program.

3.3 Experimental Procedure

In the initial experimental phase, parameters with the potential to affect weeping will be screened to identify those which are important, and to approximate their overall effects. Testing will continue until weeping is established, the field of parameters narrowed, and an accurate model developed.

Based on the information presented above, a list of parameters to be considered for examination was developed which includes the following:

Material. This allows the observation of effects resulting from material properties such as surface wetting, corrosion, and passivation tendencies.

Surface finish. Porosity, roughness and lay of marks as a result of fabrication techniques could influence corrosion and passivation layer formation, as well as provide a "hide-out" for contaminants.

Temperature. Expansion, due to elevated temperature or gradients produced by interior sources and varying ambient temperatures, could induce stresses which crack or spall the passivation layer.

Humidity. Humidity provides a potential vehicle by which contamination is transported to and from pores and cracks in the cask surface. Humidity may also act as an electrolyte in the corrosion process.

Stress. Stress results in mechanical strain, which may change grain boundaries and sizes, and possibly induce cracking. Energy differences and mechanical bonding may result at these sites.

Duration. Cask submersion time could affect the extent of surface adsorption and depth of diffusion.

Repetition (Aging). Surface voids and binding locations may be filled by cyclic exposure to contaminants and the environment over time. Together with growth and removal of oxide layers, aging may result in increased fixed contamination levels, increasing the availability of contaminants for release.

Pool condition. The parameters of a reactor spent fuel storage pool such as temperature, contaminant concentration and chemical form, and pH could conceivably affect reaction of contaminants with cask surfaces.

Decontamination. The decontamination procedure could determine the percent of smearable and fixed contamination removed, possibly exposing a layer of contamination with the potential to weep.

Vibration. Kinetic energy transferred to the surface during tranport may be sufficient to free loosely-bound particles.

The first goal of the experimental program is to induce weeping. Once weeping has been established, parameter effects may then be studied. Statistical scoping designs were selected as a systematic means of determining main parametric effects as well as limited parameter interaction effects.

The six specific parameters selected for the initial scoping tests are material, surface finish, sample and ambient temperatures, humidity, applied stress, and duration of submersion. These represent the cask surface and the primary environmental influences to which it is exposed. Repetition effects can be observed with further cycling of the same samples.

During the scoping tests, decontamination will be held to one mild method. Spent-fuel pool conditions will simply be monitored, due to the difficulty in varying temperature, contaminant concentration and pH to experimental specification in a large commercial pool. Finally, since weeping has been observed on casks in storage, vibrational testing will be deferred.

The first round of experiments consists of 122 cylinders of 304 stainless steel and commercially pure titanium. Each sample is rolled and welded, forming a cylinder. This configuration has the following advantages: a) a configuration and stress state similar to exterior sheathing in previous and future cask designs; b) a weld within each material and finish for examination of welding effects; and c) interior temperature control independent of ambient temperature for gradient establishment.

The prepared samples will be submerged in the Callaway spent fuel pool for a 7, 19, or 30 day period. Pool water samples will be taken periodically from the volume near the samples and analyzed for contaminant identity and activity. Following submergence, all sample surfaces will be decontaminated to a level equal to or below 2200 dpm/100 cm^2 (0.37 Bq/cm^2) by means of deionized water wash and wiping dry, in accordance with transportation requirements [1,2].

Prior to exposure to controlled environments, each sample will be swiped for removable contamination. Five swipes will be taken of the: 1) full area of a machined flat; 2) full area of welded and machined flat; 3) remaining welded exterior area exclusive of flats; 4) remaining exterior surface area exclusive of welds, flats, and edges; 5) interior surface opposite 4). This results in five different sets of data, allowing additional examination of the effects of welding and machining techniques on weeping. To simulate aging effects, or multiple exposures to contaminated pool water over a long term, samples may be cycled through pool submersion, environmental exposure and decontamination many times.

During environmental testing, surface temperatures of different groups of samples will be held at 0°, 100°, and 200° F, while ambient temperatures are controlled at 0°, 50° and 100° F and relative humidity held at 10%, 50%, and 90%. Each group will be exposed to 48 hours of environmental conditions, after which the swiping procedure will be repeated.

Upon completion of the scoping test series, the resulting data will be evaluated statistically to determine the main parametric effects, interactions between any 2 parameters, and indications of 3-variable interactions. These results will be compared to the physical model of weeping, which may suggest modification of the model and/or further testing.

SUMMARY AND CONCLUSIONS

Contamination weeping on transport casks is a recurring problem, despite diligent pre-transport decontamination efforts. A program is underway to investigate the contamination weeping phenomenon, and recommend design and/or operational constraints to reduce occurrences of weeping and maintain the lowest turnaround time and worker exposure.

Analysis of transportation campaign data indicates a relationship between rising levels of removable contamination and low ambient temperatures in transit. The same analysis suggests more data are necessary to explain relationships between weeping and the transporting and receiving sites. Such data includes pool water chemistry, decontamination techniques, and time the surface is in the contaminating environment.

Casks in storage have exhibited weeping, and the weeping surfaces have been replicated for SEM analysis. Visually, the surfaces as replicated are inconclusive as to structural effects. The surfaces exhibit a wide variety of characteristics from distinct grain structures to gross and fine striations with no visible grain structure. Replicated surfaces contained minerals common to surface films and dirt. Distinctions between the weeping and non-weeping surfaces are difficult to draw from this limited analysis. Additional replications and refined procedures could help clarify surface characteristics common to weeping.

A physical model of contamination weeping is under development. The model suggests that a passivation layer, which forms on stainless steel surfaces to inhibit corrosion, plays a key role in contamination weeping. During layer formation, contaminants may become incorporated into the layer structure. As the layer thickens, outer reaches of the layer may be broken away. Additionally, mechanical and thermal stresses encountered during routine cask service may strain and crack the passivation layer, allowing passage of contaminants to and from the surface.

Finally, experimental tests are underway to induce weeping under controlled conditions, clarify observed parameter relationships, and assist in the development and verification of the physical model. The first experiment utilizes 304 stainless steel and titanium cylinders totalling approximately 10 m^2 of surface area, with milled, turned, polished, and unfinished surfaces. Samples will be subjected to stress levels ranging from zero to yield strength in the course of the tests. The samples will be contaminated in a spent fuel pool, decontaminated, and subjected to temperature and humidity conditions similar to those encountered in transit. Removable contamination will be measured periodically to determine if and the extent to which weeping has occurred.

Little data could be collected on the spent fuel pool water chemistries for past transportation campaigns, due in part to utility reluctance to provide such data. Renewed efforts to secure utility cooperation should be made, particularly regarding pool chemistry, and detailed descriptions of decontamination and measurement procedures. Future experiments should include the effects of pool water chemistry and decontamination methods.

REFERENCES

[1] U.S. Code of Federal Regulations, Title 49, Part 173.443, Contamination Control, Washington, D.C., July 1983.

[2] IAEA Safety Series No. 6, International Atomic Energy Agency, Vienna, Austria, 1985.

[3] Grella, A.W., "Compliance Inspections by USNRC of Recent Spent Fuel Shipments," Proceedings of the Symposium on Waste Management, Tuscon, AZ, March 1987, pp.431-435.

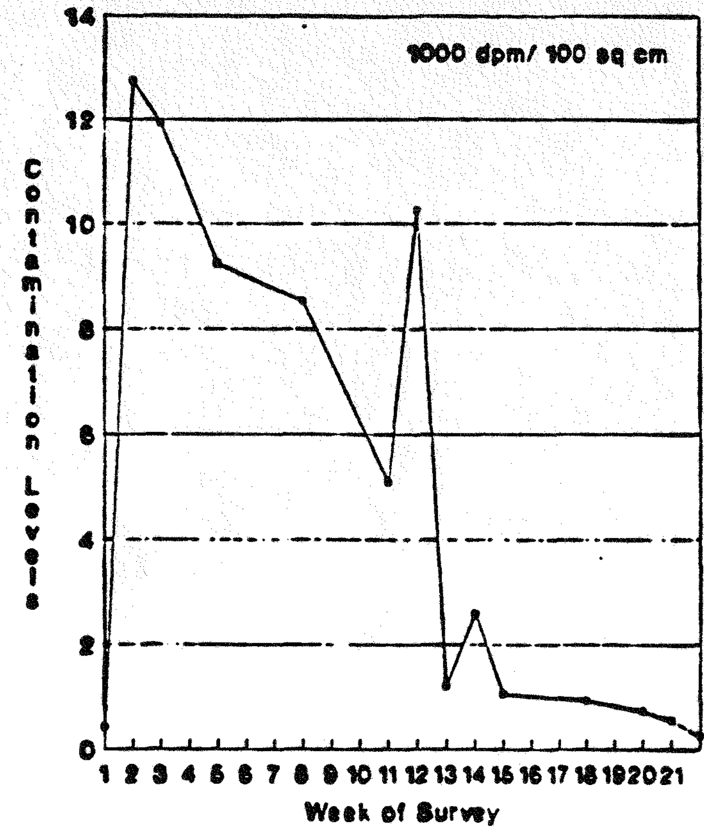

Figure 1. Smear Results - TN-8L.4

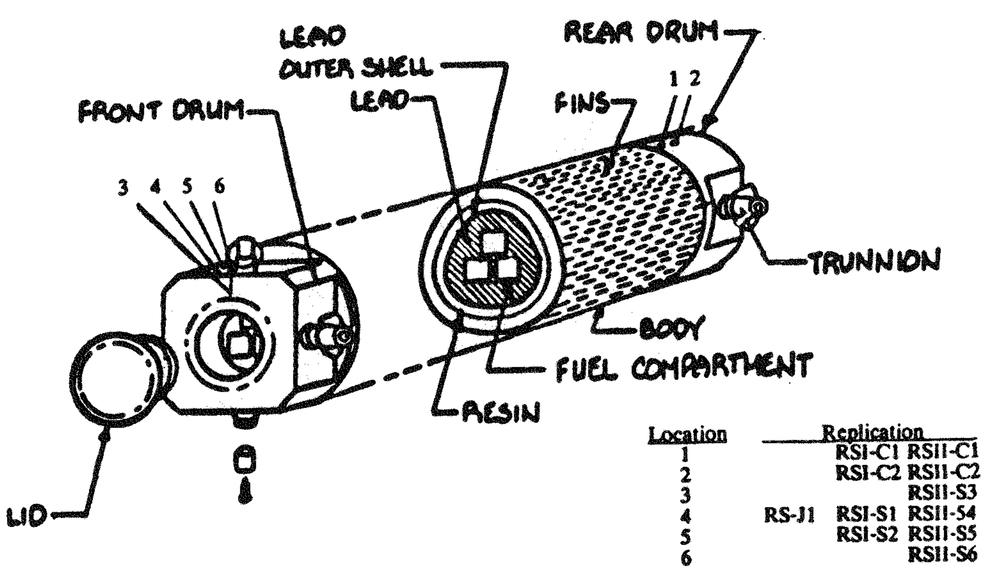

Figure 2. TN-8L.3 Replication Locations

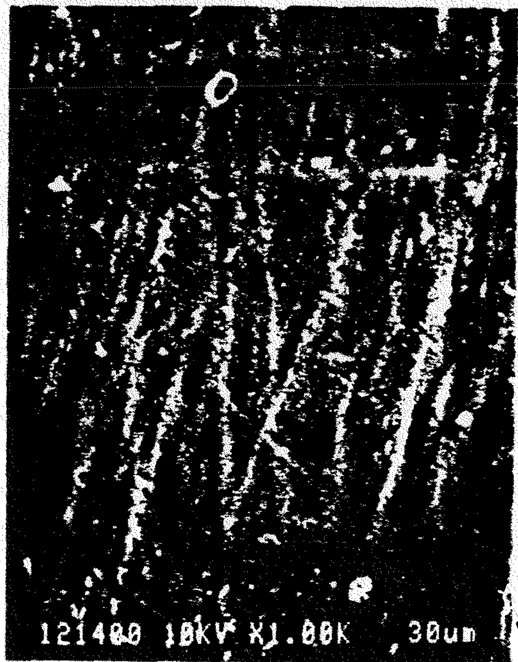

Figure 3. RSI-C1 Replication, Non-weeping Surface

Figure 4. RSI-S2 Replication Weeping Surface

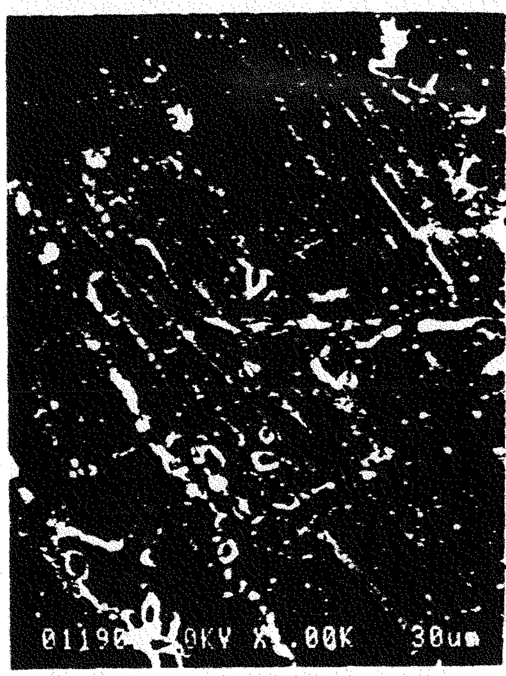

Figure 5. RSII-S4 Replication Weeping Surface

Figure 6. RSII-C2 Replication Non-weeping Surface