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In-Service Analysis of Cask Contamination Weeping*

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INTRODUCTION

Spent fuel transportation casks have arrived at final destinations with removable surface contamination levels in excess of regulatory limits (U.S. Code of Federal Regulations, Title 49 Part 173, and IAEA Safety Series No. 6), although pretransport surveys indicated removable contamination levels were well below these limits. This rise in removable contamination is called weeping. Weeping, also known as sweating, is the transformation of fixed radioactive particulates on an exterior surface of a transport cask to a removable state, some time after the cask has been removed from a spent fuel pool and decontaminated.

The weeping phenomenon is countered by time-consuming operational constraints and procedures which have a significant, adverse impact on cask turnaround times and occupational exposures at originating facilities. Furthermore, though not a significant health hazard, the arrival of a weeping cask results in negative public perceptions that are inconsistent with industry and regulatory 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 techniques
- 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.

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The Investigation of Cask Contamination Weeping at Sandia National Laboratories consists of three elements:

- Collection and analysis of in-service data
- Physical model development
- Experimental evaluation of the model and observed correlations.

This paper describes the in-service data collection and analysis and suggests direction for physical modelling and experimental evaluation.

BACKGROUND

Compilation and evaluation of data from previous transports was used to help determine, where possible, which parameters contribute to the initiation, rate, and continuation of the weeping phenomenon. Transportation records have been compiled and current transportation campaigns have been monitored by Transnuclear, Inc. (TN), General Nuclear Systems, Inc. (GNSI) and Sandia National Laboratories (SNL).

The Radioactive Materials Incident Reporting (RMIR) database maintains publicly available reports about radioactive materials transportation incidents since 1971. These were drawn from the Hazardous Materials Incident Report System of the U.S. Department of Transportation (DOT), from files of the U.S. Nuclear Regulatory Commission (NRC), and from state radiological control offices. Forty-six cases of excessive removable surface contamination upon arrival from January 1977 through July 1986 are listed in RMIR. These were all pool-loaded Type B casks, carrying spent fuel, irradiated hardware or empty, with surface contamination exceeding the 22,000 dpm/100 cm^2 limit.

Experience with recurring weeping episodes has led the NRC to term the phenomenon "chronic" (Grella 1987). From 1983-1987, NRC inspectors, routinely engaged in inspection of spent fuel transports, 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 to exceed limits after transport, even though limits were not exceeded prior to transport.

CAMPAIGN OBSERVATIONS

GNSI reported on 26 activities since 1986 involving loading and transportation of spent fuel and irradiated hardware. The reported transports used four cask models: CNS 1-13G, which is a 304 stainless steel, 20 year old cask; CNS 3-55-1 and -2, also 304 stainless steel and 25 years old; Castor V/21, which are new nodular cast iron storage casks with painted exterior surfaces; and Castor IIb, a nodular cast iron, painted transport cask used in the Federal Republic of Germany (FRG).

Two transports of irradiated hardware were made from the Big Rock Point power plant to the Barnwell Low Level Waste Management Facility in March and April 1988, using the CNS 1-13G. On receipt of the second transport, the cask exhibited a permissible level of removable contamination of 10926 dpm/100cm^2.

Eight transports of irradiated hardware were made to Barnwell from five reactor sites in the CNS 3-55-1 during the period of August 1987 to February 1989. Upon arrival of one transport, the cask displayed a level of 10000 dpm/100cm^2 at the cask lid on the weld, while the cask arrived a second time with removable contamination ranging from 14000 to 61473 dpm/100cm^2 on the cask sides and end.

Nine irradiated hardware transports from six reactor sites were carried out in the CNS 3-55-2 in the same time period. On one occasion, the cask area in contact with the spent fuel pool floor displayed 11680 dpm/100cm^2 levels. Measurements after another transport ranged from 36381 dpm/100cm^2 on the end to 122135 dpm/100cm^2 on the lid. It was noted that the highest removable contamination levels from each of the 3-55 casks occurred after transports from the same reactor site.

After such high levels were reached, a decontamination procedure using citric acid solution was carried out on both 3-55 casks. The 3-55-1 cask returned to service and experienced no further contamination problems. The 3-55-2 returned to service, and was placed in the storage yard with the sunshade on for observation after the second transport. Removable contamination levels on the lid were observed to rise to 29226 dpm/100cm^2 four days after the second transport was made. Forty days after the second transport, levels ranged from 11591 dpm/100cm^2 on the lid to 80885 dpm/100cm^2 on the side. The cask was then decontaminated, and the two following transports executed without further observed weeping.

Experience with painted surfaces was provided when six Castor V/21 casks were loaded with spent PWR fuel at the Virginia Power's Surry power plant (Table I). Each cask was originally painted with WEGOPOX 310, a green polyurethane paint made in FRG. After a loading dry run, persistently high removable contamination levels and paint residues remaining on decontamination materials prompted a recoating of cask surfaces with a hard finish white epoxy paint (Keeler and Long E-1-7475).

TABLE I. Castor V/21 Survey Summary

Cask	Surface	Hours in Pool	Survey No.	Time Btwn Surveys (Days)	Low/High Value (dpm/100cm^2)
004	Poly Paint	8	1		ALL < 1K
Dry			2	10	0 - 8K
Run			3	1	1 Pt > 1K
			4	3	ALL < 1K
005	Epoxy over	408	1		30K - 2800K
	Poly Paint		2	32	1K - 6K
			3	7	ALL < 1K
			4	7	7 Pts > 1K
	2-Epoxy over		1		ALL < 1K
	Poly Paint				
003	Epoxy over	10	1		2K - 34K
	Poly Paint		2	2	1K - 140K
			3	1	1K - 120K
			4	1	1K - 30K
			5	1	ALL < 1K
004	Epoxy over	10	1		1.2K - 28K
	Poly Paint		2	1	1K - 480K
			3	6	Up to 846K
			4	7	ALL < 1K
002	Epoxy over	10	1		ALL < 1K
	Poly Paint				
006	Epoxy over	10	1		ALL < 1K
	Poly Paint				
007	Epoxy over	10	1		1K - 350K
	Poly Paint		2	0.5	1K - 6K
			3	4	ALL > 1K

The first loading after the recoating was delayed by a dislodged seal and scheduling problems due to refueling activities, and the cask remained submerged 17 days. The refueling operation during this time may have raised the level of contaminants suspended in the pool water significantly, but sample data were not available for verification. Upon removal from the pool, there were still difficulties in decontamination, even with the new epoxy surface. Two new epoxy coats were applied, which succeded in suppressing the contaminants. The second, third and sixth casks to be loaded required multiple decontamination efforts due to rising removable levels. Monitoring at the storage location indicates the casks have remained within acceptable limits.

Similar difficulties have been experienced with painted ferritic steel surfaces in the United Kingdom. A research effort culminated in the development of a special layered paint, CEGB System 6, which was formulated for minimum contamination absorption, maximum resistance to wear and weathering, and ease of decontamination (Sanders et al. 1989).

A large number of transports by Transnuclear provided a basis for analysis of the effect of weather on the occurrence of weeping. From December, 1983, to June, 1986, TN carried out two campaigns in which spent fuel was moved in the new TN 9.1 and 9.2 casks from the West Valley Demonstration Project; 30 BWR transports were made to the Dresden power station, and 32 BWR transports to the Oyster Creek power station.

Dates, routings, and contamination levels were available from the TN campaigns. Weather records were obtained for four points along each route, and a weather profile reconstructed for each transport. Daily records included high and low temperatures, average dewpoint (humidity), and precipitation. Regression analysis was performed by SNL on the data thus collected, which resulted 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 transporting and receiving sites, H and D are the daily high temperature and daily temperature differential (high - low) which the cask experienced, β_1 and β_2 are weighting factors, and ε is the error associated with the analysis.

The site-dependent term A appears to dominate contamination levels for the TN campaign 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 temperature term appears to be a strong factor with negative coefficient, indicating that lower temperatures 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 correlation to the combined data and are not represented in the combined model. However, data from the Dresden campaign alone did indicate some relationship to the humidity and precipitation.

WEEPING ON CASKS IN STORAGE

Casks appear to undergo weeping in storage as well as in transit. Two identical Transnuclear casks, TN 8L.3 and TN 8L.4, have been under observation for weeping while in storage. Throughout the observation period, the empty casks were parked on their trailers 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 for the duration of the observations.

The TN 8L.4 cask was admitted November 5, 1987, to the TN 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.

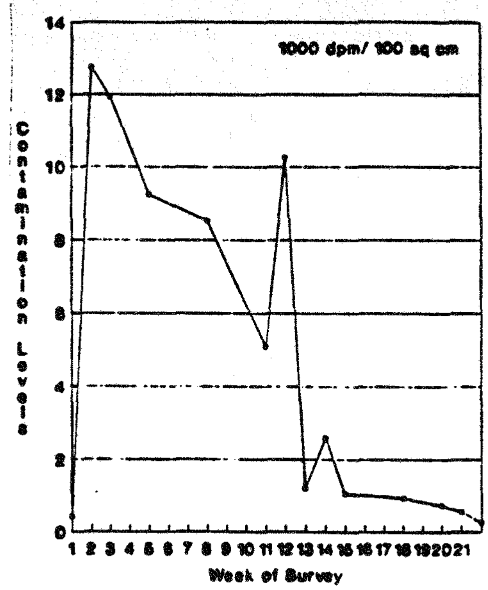
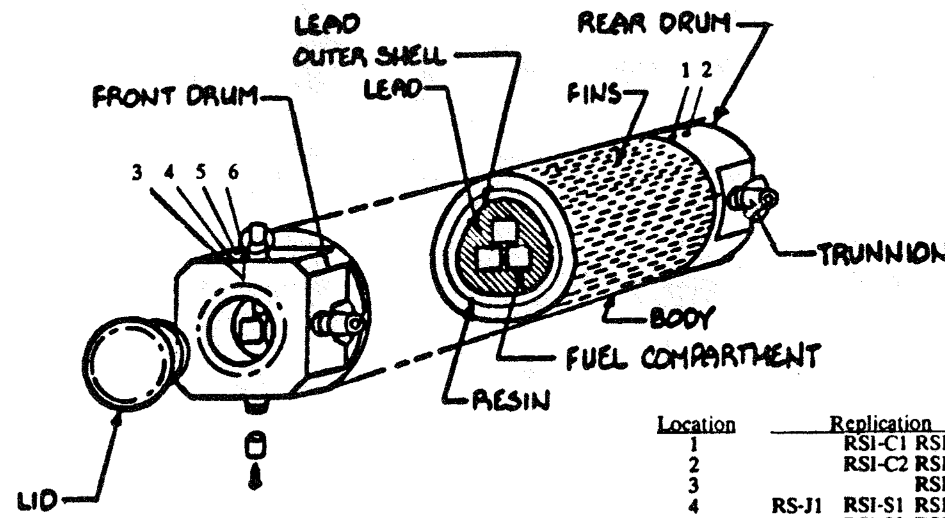


Figure 1. Smear Results - TN-8L.4

The TN 8L.3 cask completed a transfer campaign between Duke Power Company's Oconee and McGuire stations on February 19, 1988. It was admitted to the Aiken storage yard with removable contamination levels below 1000 dpm/100 cm^2. By the end of March, the level rose to 10500 dpm/100 cm^2 on the front drum. The cask was then decontaminated and used in a campaign from April 14 to May 19. The levels after check-in at Aiken remained below 1000 dpm/100 cm^2 until August, at which time they rose again to 9000. Levels fell again to near 3000 until November 28, 1988, when they reached 11000. December levels dropped to 5000 dpm/100cm^2, followed by further gradual decline.

To determine possible correlations between cask surface structure and its 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 (taken at the locations indicated in Figure 2). The films were then removed, sputtered with a Au-Ir target and examined at SNL under a Scanning Electron Microscope (SEM).

Surface replications were made at three different times: July, October, and December of 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. 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, which is a replicated weeping surface, displays a relatively clear etched grain structure.



Location		Replication	
1		RSI-C1	RSII-C1
2		RSI-C2	RSII-C2
3			RSII-S3
4	RS-J1	RSI-S1	RSII-54
5		RSI-S2	RSII-S5
6			RSII-S6

Figure 2. TN-8L.3 Replication Locations

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

Figure 4. RSI-S2 Replication, Weeping Surface

The third set of replications of the same surfaces (RSII series) was made in December. 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 II.

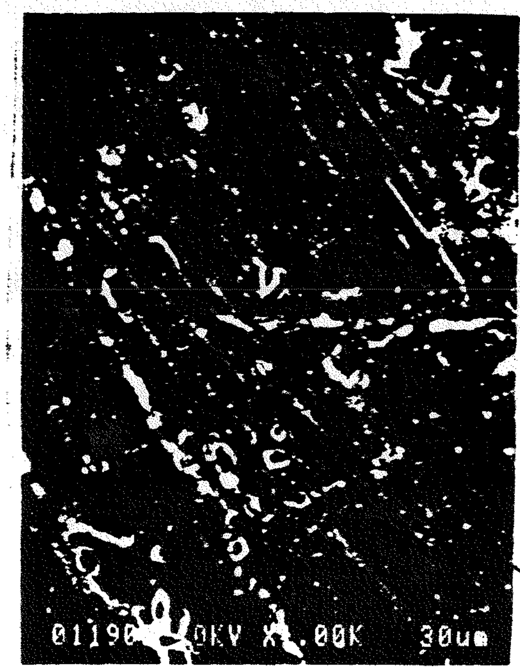


Figure 5. RSII-S4 Replication, Weeping Surface

Figure 6. RSII-C2 Replication Non-weeping Surface

TABLE II. Surface Replication Contaminants

		Contaminant (pCi)				
Sample	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

CONCLUSIONS AND RECOMMENDATIONS

Weeping occurs as a transformation of apparently fixed surface contamination to a removable form. Weeping has been observed on a variety of cask surfaces, both in transit and in storage, and on casks which were loaded or empty. Cesium 137 appears to be the primary contaminant in weeping, followed by Cobalt 60 and Cesium 134. A surface layer may be forming on the stainless steel surfaces examined obscuring grain structures while retaining or releasing contaminants.

A regression model was generated from TN transport data which indicates a correlation of low maximum ambient temperatures and low temperature differentials with weeping. Also, this model together with GNSI experience indicated more information is necessary to explain site-dependent factors.

The observations presented suggest direction for further investigation and experimental clarification. Cask submersion time could affect depth of diffusion and the extent of surface adsorption. 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. The effectiveness of a decontamination procedure could determine the percent of smearable contamination removed, and whether an area is missed altogether. The effects of weather factors should also be verified. Expansion and contraction due to changing temperature, or gradients produced by interior heat sources and varying ambient temperatures could provide mechanical release from the substrate. Humidity may provide a vehicle by which contamination is transported from pores and cracks to the surface of a cask. It may also act as an electrolyte in a corrosion process, potentially releasing fixed contamination.

Different cask surface materials and conditions could affect weeping. Material properties such as chemical bonding, surface wetting, and corrosion tendencies, as well as porosity, roughness and lay of marks as a result of fabrication techniques may influence contaminant bonding.

Operationally, increased emphasis on loading pool cleanliness, and minimization of cask immersion time in the pool would assist in the prevention of initial contamination. To assist in decontamination, cask surfaces should be maintained as smooth as possible. If painting or coating of a cask surface is required, consideration should be given to characteristics such as finish and contaminant affinity. Decontamination should be thorough, but decontamination practices which may damage the surface should be avoided. When treatment is necessary, a citric acid solution may be indicated. Finally, careful surveying of outgoing casks should be performed prior to cask release, and standardization of instruments and swiping techniques used for this purpose is recommended.

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