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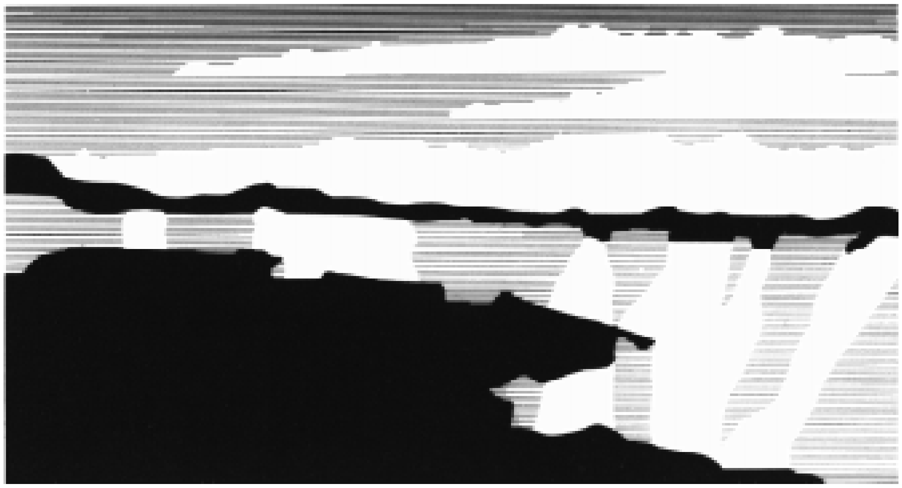

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RISK-BASED PRIORITIZATION FOR PLUTONIUM RESIDUE HOLDINGS

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1. ABSTRACT

The Los Alamos National Laboratory (LANL) has developed a risk-based prioritization methodology for plutonium residue holdings. This methodology was developed not only as a tool to assist processing personnel to prioritize the remediation of legacy materials but also to evaluate the risk impacts of schedule modifications and changes. Several key activities were undertaken in the development of this methodology. The most notable is that the risk assessments are based on statistically developed data derived from sampling and processing that explicitly measures container integrity. Also, the time-dependent behavior of these materials was modeled and included in the risk analysis. These residue items were identified in the Department of Energy's Implementation Plan for the Defense Nuclear Facilities Safety Board Recommendation 94-1. This paper summarizes the development of this methodology as well as the application of the methodology to the residue holdings at LANL.

The results show the quantitative risk reductions to vault-operations personnel that can be achieved by proper prioritization of the remediation schedule at Los Alamos. A hypothetical processing schedule that is not prioritized with respect to risk increases the overall program risk by about 50% as compared to the present schedule. The results are also used to describe the risk reductions that have been achieved by the timely implementation of remediation at Los Alamos. A hypothetical 5-year delayed schedule increases overall risk compared to the present schedule by a factor of about 2.

This methodology could be used to develop risk-based prioritization for stabilizing and processing similar materials throughout the weapons complex. Finally, since contemporary

approaches to risk management are based on static methods for estimating risk, they may inadvertently focus too heavily on the immediate risk. The result is that heavy reliance on traditional methods for risk determination may result in the elimination, stagnation, or extension of programs. These actions may result from the inherent limitations of current risk management techniques, which lead to a failure to appreciate long-term risks from postponing remediation activities.

2. INTRODUCTION

Beginning in 1993, Los Alamos National Laboratory recognized that its primary vault in the plutonium facility had nearly reached capacity. The bulk of the items in the vault contained residues that were not intended for long-term storage. To alleviate the storage problem, a program was initiated to process the residue materials, reduce the number of containers, and convert all actinide-bearing materials into a stable form, either metal or oxide. This program is known as the vault workoff program. Concurrently concern was growing about the safety of storing actinide-bearing materials at all DOE nuclear facilities. These concerns were formalized by the Defense Nuclear Facilities Safety Board (DNFSB) as expressed in their DNFSB 94-1 Recommendation. In 1994 the Department of Energy (DOE) undertook the Plutonium Vulnerability Assessment (PVA) to identify environment, safety, and health vulnerabilities arising from storage of plutonium[1]. Risks to workers were established in a very qualitative way. In February 1995 DOE issued their 94-1 Implementation Plan (94-1 IP)[2], which calls for DOE sites to complete the remediation of plutonium-bearing materials by 2002. The part of the plan pertaining to LANL is referred to as Los Alamos 94-1.

In March 1995 Los Alamos conducted an exercise using a statistically based process to select and examine a number of containers stored in the main plutonium vault. Los Alamos also undertook a 100% visual examination of all containers in the primary plutonium vault. The purpose of this sampling was to obtain information from which estimates of the risk of container failure could be developed. Failure of containers in the vault or during their movement can result in radioactive contamination of workers or the workplace. Additional inspections of statistically selected containers were performed in August 1995 and in July 1996. In addition to the body of information obtained from these inspections, the condition of containers removed from the vault for processing was recorded and included in the database of information used to calculate risk.

3. BACKGROUND

The original Los Alamos 94-1 schedule for processing was based on the assumed capacities of the various residue processing lines and qualitative information on the risk behavior of the many types of residue materials. Thus, processing experience and plant capacity greatly influenced the original processing schedule. To establish a more technical basis for the processing program a method for risk-based prioritization was developed.

Items in the plutonium facility vault containing residues are being processed or “worked-off” continuously, and new items are constantly being added to the vault. Items stored in the vault thus constitute a dynamic population. However, the 94-1 recommendation and the 94-1 IP require the work-off of legacy items stored in the vault on May 1, 1994. Even though the vault inventory is dynamic, this analysis focuses on that inventory of legacy materials and the work-off of that inventory.

The workoff program has progressed for over two years. The items that have been processed to a stable oxide form in the first two years of the program are listed in Table 1. Table 2 lists the remaining inventory in the Los Alamos 94-1 program. This inventory listing has had all of the redundant items, such as duplicate entries for a single item arising from multiple material types in that item, removed. For example, a single item may be identified by both a material type for plutonium content and a material type for uranium content. Also, we have removed from this listing materials required for use in DOE programs.

Table 1 Total Items Worked Off After Two Years on the Los Alamos 94-1 Program

	FY95		FY96		Totals	
Category	Projected	Completed	Projected	Completed	Projected	Completed
High-priority compounds	45	20	0	0	45	20
Compounds	13	58	0	135	13	193
Gases	0	0	1	1	1	1
Combustibles	10	113	0	84	10	197
Solutions	0	66	250	286	250	352
Metal	265	372	200	233	465	605
Noncombustibles	13	26	0	63	13	89
High-priority residues	224	111	250	340	474	451
Process residues	20	94	0	37	20	131
Totals	590	860	701	1179	1291	2039

Table 2 Remaining Los Alamos 94-1 IP Inventory by Detailed Material Category

Impure Metal:	**453**	**Combustibles:**	**95**	**Pure Metal:**	**322**
Other Impure Metal	386	cellulose rags	67	>100 grams	322
Spec Alloy	67	paper/wood	21		
		other	7		
High-Priority Residues:	**777**	**Process Residues:**	**983**	**Oxides:**	**529**
calcium metal	3	calcium salt	13	>100 grams	529
evaporator bottoms	3	CaO	1		
hydrogenous salt	3	cement powder	1		
filter residue	234	cemented in drum	2		
hydroxide precipitate	132	incinerator ash	117		
sweepings/screenings	294	salt	849		
oxalate precipitate	1	sample residue	0		
Sand, Slag, and Crucible	75				
silica	32				
Noncombustibles:	**575**	**High-Priority Compounds:**	**58**	**Compounds:**	**840**
asbestos	1	acetate	0	<100 grams	
filter media	1	chloride	15	PuO_2	680
fire brick	1	fluoride	0	Oxide items	2
glass	10	hydride	3	U_3O_8	54
graphite	60	nitrate	2	carbide	99
heating mantle	2	phosphate	0	nitride	5
HEPA filters	14	sulfate	3		
leaded gloves	4	tetrafluoride	26		
MgO	209	trichloride	2		
nonactinide metal	199	other	7		
plastic/Kimwipes®	51				
ion exchange resin	4				
rubber	4			**Sample Returns**	**149**
other	15				
				Totals =	**4781**

4. INSPECTION DATA ANALYSIS

4.1. CONTEXT OF RISK METHODOLOGY

The focus of the analysis is not to determine the actual risk levels associated with the Los Alamos 94-1 program, but to assign relative risk levels associated with the different categories of materials that are to be remediated as part of the program. Critical to the analysis is a direct link between observed characteristics of stored items and the risk posed by these items.

During the nearly 20 years of vault operations at the plutonium facility, no containers of fissile material have failed in an uncontrolled environment. Nuclear materials are stored in a cold vault, where a container failure could induce substantial costs of cleanup as well as a potential for worker health risks. Most fissile material is stored in containers with at least three barriers. The typical container configuration is an inner can (which is contaminated from the glovebox environment), a bagout bag (which is contaminated on the inside but is clean on the outside), and an outer can (which is clean on both the inside and outside). Although the inner can is contaminated, it confines the bulk of the residue material, and its failure is coupled to failure of the bagout bag, which cannot continue to confine the residue for very long after failure of the inner container. Thus, the inner container and the bagout bag function are characterized as a single confinement barrier in our analyses. While many variables affect the ability of the inner container/bagout bag combination to contain the fissile material, only two strong discriminators have been identified: matrix material and origin of the item.

Physical processes in the inner container/bagout bag dominate the degradation mechanisms. These processes are, in turn, governed principally by the matrix material contained. This was one of the reasons for maintaining a database that included matrix material as an entry. Items that originated from the facility performing many of the chemical analyses, in general, were packaged with a different bagout bag procedure. Also, these items tended to be smaller, less stable for a given matrix material, and were generally packaged with thinner bagout bags. These items demonstrated a significantly higher probability of failure of the inner container/bagout bag than the bulk of items in the plutonium facility vault.

4.2. NATURE OF INSPECTION DATA

The inspection data used in this analysis are from one of three separate sampling exercises or from the processing of items. Shortly after the March 1995 sampling program, an inspection program was instituted requiring all processed items to undergo the same inspection as those items in the sampling program. This inspection program has generated a large amount of data that complements the data from the sampling exercises. Since many of the items being processed are in the high-risk category, much of the inspection data acquired applies to residues in this category.

Plutonium is packaged in several ways, but most of the items considered here are placed in an inner container or "can," then the can is placed inside of a plastic bagout bag, which is twisted shut and tied or taped. There may be two of these bags. Finally, the bagged item is placed inside an outer container or can. When items are inspected, they are first screened visually for damage to the outer container. Then the inside surface of the outer container is "wiped" to detect contamination. If the inner surface of the outer container is contaminated, the bagout bags or inner container are characterized as "failed" (failed to provide a contamination barrier) and further information on the mode of failure is sought. The inner and outer cans may fail from corrosion. The bagout bags may fail by becoming untaped, worn from the weight of the inner can, torn from sharp edges, or degraded from radiolytic damage. In the latter case, they become blackened and brittle.

When a container fails the screening criterion (denoted as a "failed container"), the date the container failed is not known; we know only that it failed some time after the packaging (or "creation") date and before the inspection date. Similarly, for items that pass the screening criterion, we know only that they survived from the date of creation to the date of inspection. In most reliability studies, the actual lifetimes of the failed units are available for analysis. In our case, we have only "interval" data, which introduces a much larger uncertainty into the results.

The unfailed units are said to be “censored on the right,” meaning that their lifetimes are not known.

4.3. VISUAL INSPECTION

The 100% visual inspection of the PF-4 vault was performed over a three-workday period and utilized a total of 32 employees. This inspection was completed 1.5 months ahead of the schedule to support the inventory risk assessment.[3] This exercise, while valuable for determining the present condition of exterior containers in the vault, came at considerable operational expense. The average dose in the vault is approximately 35 mrem/hour, and each individual was constrained to a maximum of two hours of operations. This inspection required about 70 man-hours and resulted in a cumulative exposure of approximately 2.2 rem. Further information on the 100% visual inspection is available from internal documentation.[4]

The results of the visual inspection of the PF-4 vault were that 361 containers out of 5876 were found with some visually observable defect. (The total number of containers inspected, 5876, differs from the number of items listed above because a container may have more than one item in it. The term container represents a single outer unit.) Of these, 82 containers appeared to have lost primary containment as evidenced by raised lids, corrosion, or other factors. This indicates that presently 1.4% of all items have an outer barrier that is no longer intact and sufficient to contain the spread of radionuclides. This information was used to provide a basis for the probability of failure of the outer container. Though a significant number of containers failed the screening criteria at Class 1, no external contamination was observed. This indicates that no containers in which both barriers are breached presently exist in the vault.

4.4. OVERALL INSPECTION RESULTS

Table 3 lists the average fraction of items in these categories that were found to fail the screening criteria as well as the 90% confidence bound on that fraction. The 90% confidence bound reflects the expected upper limit of the failed fraction if the entire inspection database were to be collected 10 independent times. (More precisely, if the entire database were to be collected a very large number of times, the 90% confidence bound represents the upper limit on 10% of those collections.) Table 3 is critical to our analysis in that it completely describes the state of the legacy items as they exist at the time of the inspection. With the large number of items presently in the inspection database, the fraction of remaining items that are likely to have an inner barrier that fails the screening criteria in the near future is well understood. The full details of the specific items that failed the screening criteria are available elsewhere[5,6,7].

Table 3. Average Fraction of Items that Failed the Screening Criteria and the Estimated Failed Fraction at 90% Confidence

Material Category	Fraction of items that are expected to have an inner barrier failure	90% confidence limit on the fraction of items that are expected to have an inner barrier failure
Oxides	1.0%	3.9%
Pure Metal	1.9%	3.5%
Sample Returns	18.0%	20.0%
Compounds and Noncombustibles	1.7%	3.3%
Impure Metal	1.7%	3.2%
Process Residues	2.6%	3.1%
High-Priority Materials	7.0%	9.6%

A graphical representation of the inspection results is useful in interpreting overall inner barrier failure rates as well as age-specific analysis. In Figs. 1–3, the overall inspection data results for three of the material categories are shown.

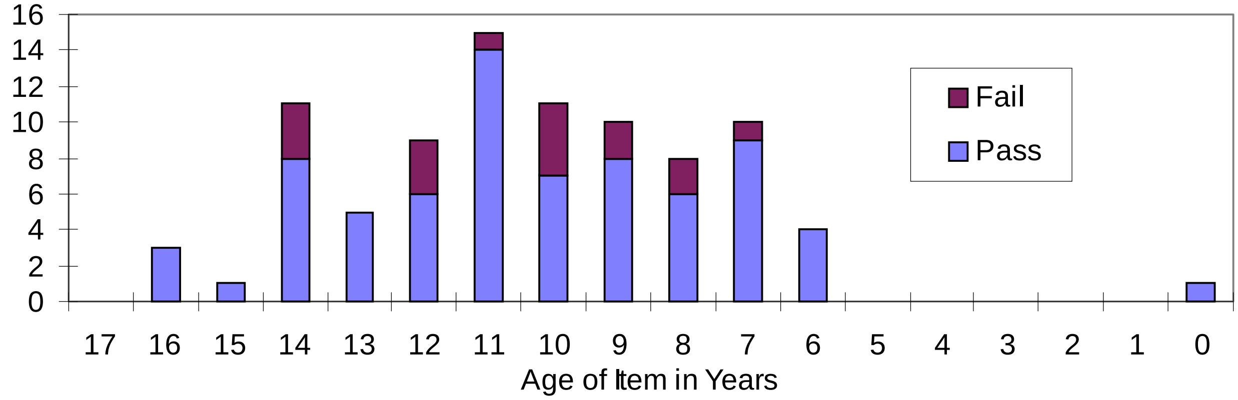

Figure 1. Results of all inspection data for the sample return items.

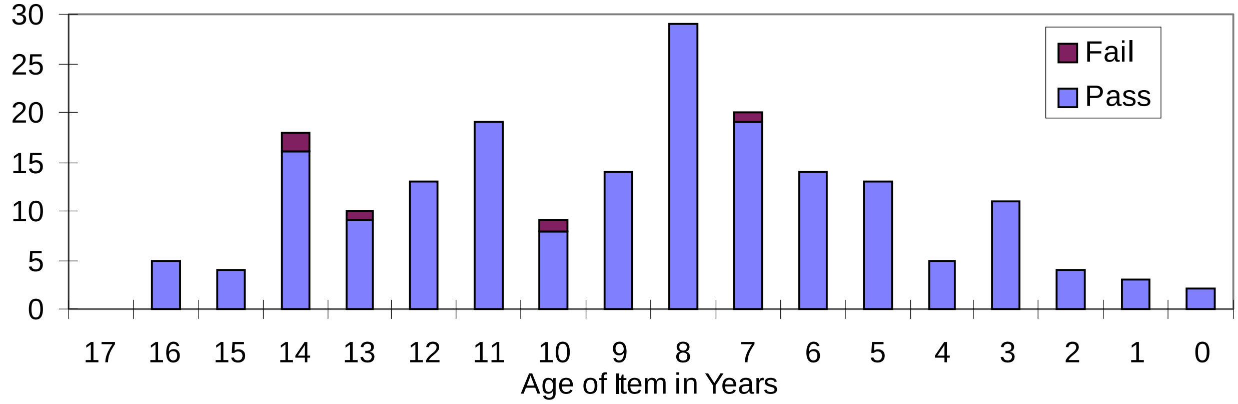

Figure 2. Results of all inspection data for the process residue items.

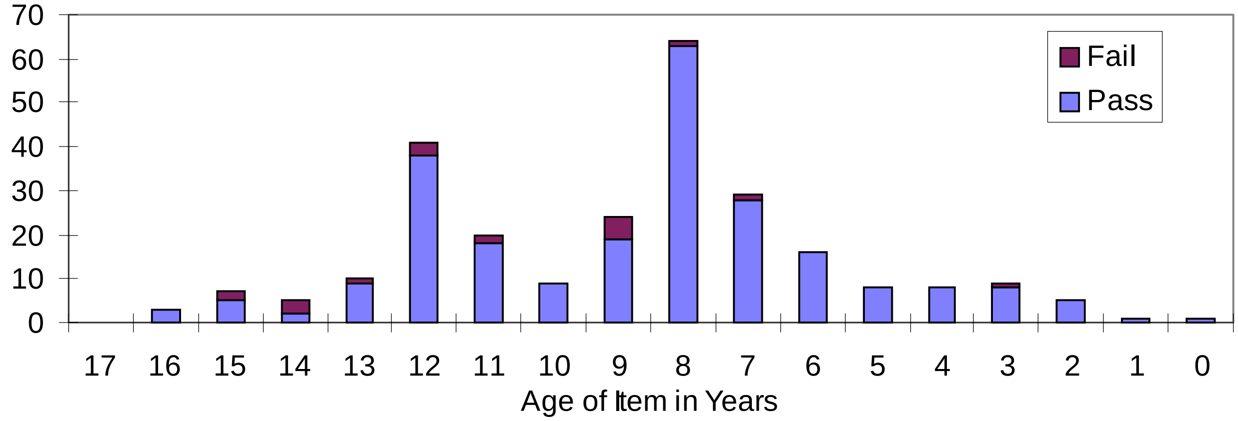

Figure 3. Results of all inspection data for the high-priority items.

4.5. MODELING LIFETIMES OF CONTAINERS

4.5.1. Limits of Traditional Approaches

The above analysis provides estimates as to the present condition of the vault inventory. The PVA suggested that a significant fraction of the present inventory may be in a deteriorated state. This qualitative conclusion from the vulnerability assessment is not supported by the present data across the whole inventory at Los Alamos. However, for those items known to be high risk, that is., High-Priority Process Residues, the fraction failing the inspection criteria is significantly higher than the average. Another significant risk factor outlined in the PVA is the potential failure rate of containers. The PVA suggested that certain categories of materials, particularly high-risk materials and plutonium-bearing salts, may be susceptible to a “light-bulb” effect. In other words, these items would pass a screening test now, but may collectively fail as a category with a very high rate after some “latent” period. This behavior can be characterized by a Weibull distribution with a large shape factor; e.g., $\beta = 8$.

4.5.2. Historical Experience

Essentially all objects, from airplanes to electronics, exhibit failure characteristics that can be described by the so-called “bathtub” curve, which is comprised of the three types of failures shown in Fig. 4. As an example, initial failures in electronic components tend to be large compared to the probability of chance failure within a specified time, but chance failures and failures from aging also occur. This approach can be applied to the material categories for the legacy plutonium, that is., the seven categories used in this analysis.

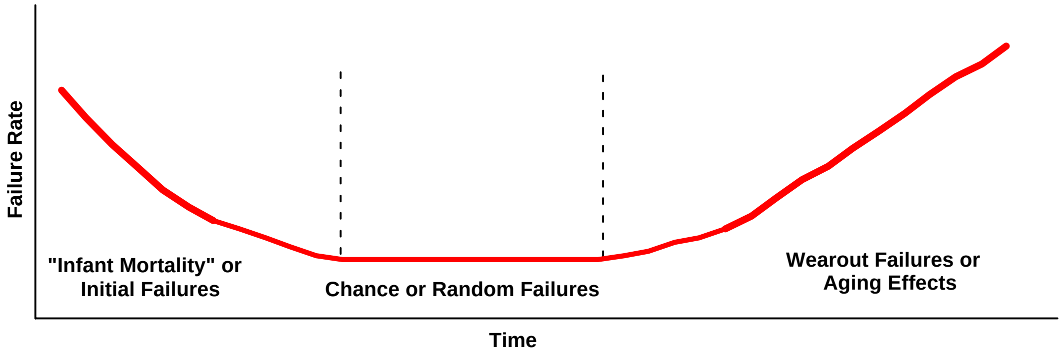

Figure 4 Typical “bathtub” curve representing reliability characteristics of engineered systems.

Historical complex-wide experience demonstrates that plutonium oxides can be packaged adequately for long-term (>50 y) storage. These materials when heated to a low loss-on-ignition condition have little driving force to another, lower energy, state. However, incorrect or incomplete packaging procedures have produced failures in containers with plutonium oxides. The material characteristics lead to the conclusion that the breach of plutonium oxide containers will be dominated by an “initial failure” type of reliability characteristic. This initial failure characteristic is commonly modeled by a Weibull distribution with a shape factor less than 1. The particular value (<1) chosen for the shape factor becomes unimportant under the assumption that failures in the time frame of interest are dominated by initial causes. In the analysis of the plutonium oxides, it was assumed that any failures have already occurred. In other words, no new failures will occur in time.

In contrast to oxides, other plutonium materials have demonstrated aging characteristics. These include metals and process residues, including plutonium salts. These materials are characterized by the upward sloping portion of the bathtub curve. Typically, the reliability of these types of objects is modeled by a Weibull distribution with a shape factor greater than 1. When the shape factor is greater than 1, the object has an increasing failure rate. If the shape factor is equal to 2, the failure rate is linearly increasing; this is a special case of the Weibull, called the Rayleigh distribution. When the shape factor is greater than 2, the objects have super-linearly increasing failure rates.

Between these extremes lie a large fraction of the materials that tend to demonstrate no particular aging phenomena. The behavior of these materials may be dominated by packaging problems. In fact, analysis of the cause of failure for the 22 items found in the two random sampling exercises suggests that packaging-related problems may dominate. If the failure of a bagout bag is related to the physical abrasion encountered between the bag and the inner or outer container, the failure rate is expected to be constant in time. This is because the containers are handled at a relatively constant rate for materials-accountability requirements. Furthermore, complex-wide experience has not shown any known peculiar behavior for these types of materials. In general, container failures have been rather random or chance dominated.

4.5.3. Aging Model

Using the lifetimes (or approximate lifetimes in our case) of inspected units, we would like to predict the lifetimes of the units that were not inspected. This predictive ability allows us to calculate risk of container failure (as a function of age and category of material of the units) and to relate that risk to the risk of contaminating workers or the workplace. Prediction requires a model for lifetimes. We chose a flexible and widely used model, the two-parameter Weibull Distribution, which includes, as a special case, the exponential distribution. The density function of the Weibull is

$$f(y) = (\beta / \alpha^{\beta}) y^{\beta-1} \exp(-(y/\alpha)^{\beta}) \text{, where } y>0. \qquad (1)$$

The parameter α is the *scale* parameter, and β is the *shape* parameter. The parameter α represents the spread of the distribution; in particular, it is the 63rd percentile of any Weibull. The parameter β represents the shape of the distribution: for β =1, the distribution drops off exponentially; for values of β between 1 and 3, the distribution is unimodal and skewed to the right; for values between 3 and 4, the shape resembles the normal distribution. More importantly, the Weibull hazard function, which represents the instantaneous failure rate at age y, is constant for $\beta = 1$ (not a function of age) and is an increasing function of age for higher values of β. When $\beta = 2$, the hazard function is linearly increasing, and the distribution becomes the Rayleigh distribution.

The items in the vault were created or packaged at various times. (Most items are “created” when the material is packaged into a new container, but this may not be the case in all instances.) To analyze the lifetimes, we consider that all the items are created at time zero and subtract the creation date from inspection date. The “new” inspection date of the i-th unit then becomes d_i = (actual inspection date – creation date). In our sample, m of the items fail the screening criteria before inspection (have lifetimes less than d_i), and the others are “censored” (have lifetimes longer than d_i).

To apply the maximum-likelihood technique, we will assume that the lifetimes of all items have a Weibull distribution. The probability that the lifetime is less than or equal to d_i is the cumulative Weibull,

$$F(y) = 1 - e^{-(d_i/\alpha)^{\beta}} , \tag{2}$$

and the probability that the lifetime exceeds d_i is

$$1 - F(y) = e^{-(d_i/\alpha)^{\beta}} . \tag{3}$$

The likelihood is thus

$$L = \prod_{i=1}^{m} (1 - e^{-(d_i/\alpha)^{\beta}}) \prod_{i=m+1}^{n} (1 - e^{-(d_i/\alpha)^{\beta}}) . \tag{4}$$

A simplex (Nelder-Mead) search routine was used to find the values of α and β that maximize the likelihood function; these values are called the "maximum-likelihood estimates" (MLEs). There is no need to take derivatives, but taking the log of the likelihood function simplifies the numerical problems (getting out of bounds). We obtain parameter estimates for each of the seven categories of material packages. The parameters are highly correlated and likelihood function can be very flat, so that one parameter can be changed significantly (if the other is adjusted accordingly) without making much change in the likelihood.

We now address the issue of uncertainty in the maximum-likelihood estimates of the parameters. Although there are several ways to approach this, we chose to use the likelihood ratio λ. Theoretically, $(-2 \ln \lambda)$ has a chi-square distribution with 2 degrees of freedom; that is, the 90th percentile or "critical value" is 4.605. We used a grid search to find values of α and β for which the absolute value of $\{(-2 \ln \lambda) - 4.605\}$ is small. This produces a confidence region that tends to have a long "banana" shape as shown in Fig. 10 and a corresponding vector of values of α and β.

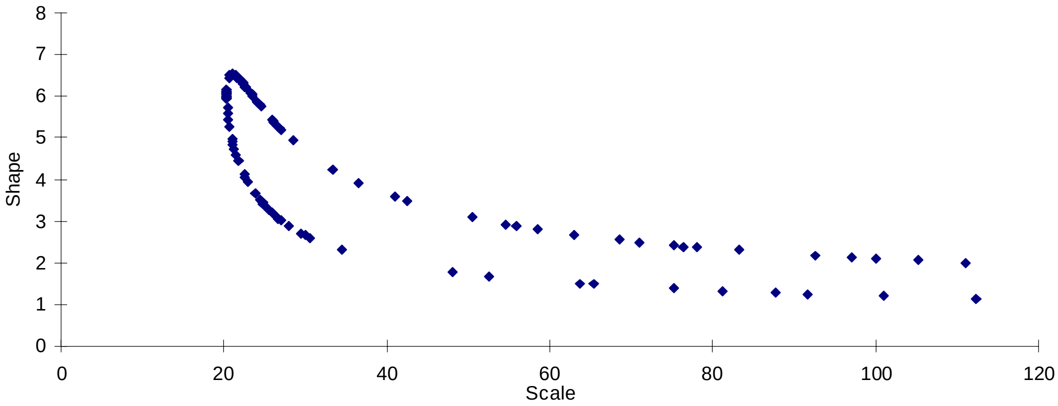

Figure 5. Ninety percent (90%) confidence region of the scale and shape parameters for the material category High-Priority Materials.

To facilitate modeling, a transformation of the parameters to $1/\alpha$ versus β gives a more ellipsoidal shape. Asymptotically, the parameter estimates are normally distributed. Next the means, standard deviation, and correlation coefficient of the vectors were obtained. We then generated observations from a bivariate normal with these means and correlation coefficients (ρ). This was done by generating two random normal numbers, y_1 and y_2, with mean of zero and unit standard deviation. The simulated values are then

$$1/\alpha = y_1\sigma_1 + \mu_1 \tag{5}$$

and

$$\beta = \left(y_1\rho + (1-\rho^2)^{1/2} y_2\right)\sigma_2 + \mu_2 . \tag{6}$$

The means and variances were adjusted so that the bivariate normal ellipse would fit within the confidence region. For each item in the database, we generated a pair of parameters from the bivariate ellipse and used them to generate a random lifetime for that unit (that is, we calculated a lifetime = $\alpha[-\ln(1-u)]^{1/\beta}$, where u is a random number from a uniform distribution). Where the expected value of β was closer to 1 but with larger uncertainty, as for oxides, pure metal, compounds and noncombustibles, and impure metal, we drew the random variables, y_1 and y_2, from the gamma distribution instead of the normal distribution. This facilitates the elimination of subzero values for $1/\alpha$. Finally, we added the simulated lifetime to the actual creation date to obtain a simulated failure date for each item. This new database is called the simulated inventory database. Using the simulated inventory database, the number of failed containers and the probability of failure can be calculated at any time. Figures 6 shows the transformed confidence regions with a 100-point simulated database generated using this technique for three material category.

While the MLE technique provides for an analytic solution to the reliability parameters as a function of material category, the parameter estimates have very large uncertainties when few failures exist per category. This leads to a situation that grossly overestimates the uncertainties in performance of the material category.

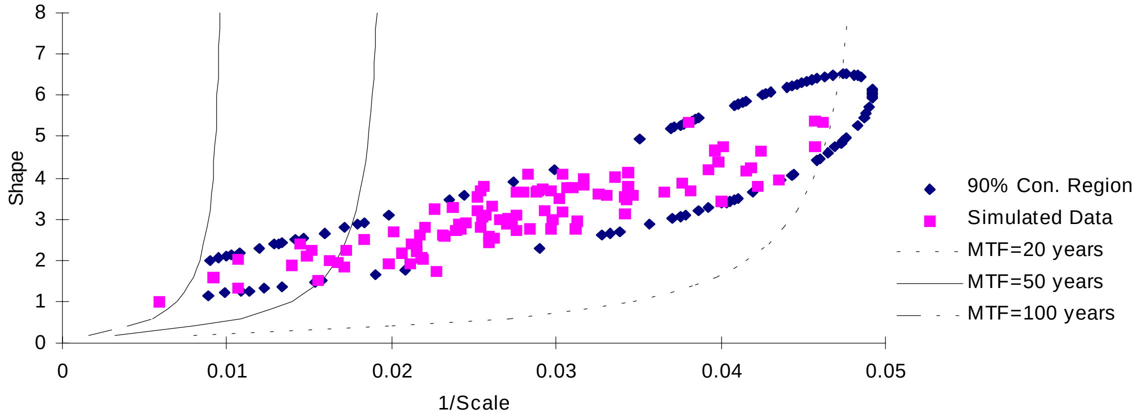

Figure 6. Confidence region (90%) for the distribution parameters for the high-priority category. Also plotted are 100 simulated data points and curves showing 20-, 50-, and 100-year mean-times-to-failure.

Another approach to evaluating the parameters of the distributions for the various material categories is to examine the failed fraction of items as a function of item age. For the Weibull distribution the age-specific failure rate is given by the hazard function:

$$h(y) = (\beta / \alpha^{\beta}) y^{\beta - 1}. \tag{7}$$

We have used a nonlinear least-squares (NLS) fitting algorithm with binomial weighting to fit the hazard function to the failure data. The results from the maximum-likelihood fit and the least-squares fit are shown in Table 4. For the maximum likelihood there is a range of values with nearly identical likelihood. We have chosen values nearest the least-squares values provided that the likelihoods were within 0.1 of the converged values.

Physical considerations can add information to the total picture. For oxides, the two failures were items packaged on the same day and with adjacent “Lot ID” numbers. This means they were not independent failures, but represent a single failure, perhaps due to improper packaging technique, but not likely an age-related or a materials-related failure. Thus, with only a single failure point we cannot analytically compute the age-related behavior for this material category. In terms of the parameters for the Weibull distribution used to characterize failure of oxide item, we must return to the “engineering approach” that suggested that a constant failure rate, $\beta = 1$, is the most likely.

For pure metals, the situation is similar. We have two failures at four and five years and only one at late times. The bulk of the metals are young in age, however. While the MLE and NLS techniques generate parameters for the distribution, they have very broad confidence bounds. This general problem also exists with the materials categories of “Compounds and Noncombustibles” and “Impure Metal.” In each case, we have very few failures and mixed results with regard to aging phenomena. Because of these issues, the shape parameter for these categories is assumed to be near 1 (constant failure rate). The actual parameters used in the failure prediction calculations are a generated by coupling this engineering approach to the results from the two analytical techniques.

Three of the material categories provide sufficient failure data to make use of the analytical techniques without reservation: sample returns, process residues, and high-priority materials. For the high-priority materials category, the failure rate appears constant until age 14. There are 5 failures out of 12 samples there, but 3 of the failures are silica solids, and 2 are rags. The July sampling uncovered the fact that these two subcategories had high failure rates. If we neglect these two small subpopulations, the failure rate appears to be quite constant. For the sample returns category, 4 of 17 failures are graphite crucibles, and exclusion of this small subpopulation would leave us with what appears to be a constant failure rate. We conclude then, that there is less of an aging effect than first appears, that is, β may be closer to 1 than shown in Table 4. In order to provide a more conservative estimate on the future number of items that may fail the inner barrier, we maintain the higher β for the analysis.

A weakness in the maximum-likelihood technique for estimating the parameters of the Weibull distribution occurs when only a few failures are observed and when the data are interval data as in this case. In short, the technique does not appear to utilize the large amount of information available from the number of passes in a given category (as opposed to the number and date of failures). As shown in Table 3, the uncertainty in the *present* configuration in the vault as described by the hypergeometric distribution is quite small for most of the material categories. Further, these numerically based techniques (either the maximum-likelihood, or the weighted least-squares fit o-n the hazard function) do not make use of the engineering perspective on physical mechanisms for failure. Thus, we arrive at a situation where some subset of the material categories is still best estimated by use of physical arguments for the β parameter and a fit to the α parameter.

Table 4. Summary of Weibull Parameters Estimated by Various Techniques and the Original Estimates

Material category	Nonlinear Weighted Least-squares Fit on Hazard Function.		Maximum-Likelihood Technique	
	Scale (α)	Shape (β)	Scale (α)	Shape (β)
Oxides	NA	NA	NA	NA
Metal	454	1.10	150.	1.29
Sample Returns	48.9	1.61	50.1	1.02
Compounds and Noncombustibles	79.8	2.24	36.5	3.12
Impure Metal	5140	0.3	1400	0.80
Process Residues	94.9	1.77	94.9	1.65
High-Priority Materials	44.8	1.76	51.1	1.57

In Table 5, we show the actual parameters used for the material category's expected performance of the inner barrier and the results of using these parameters for both the best estimate case and the 90% uncertainty bounded case. For all material categories, the uncertainty bounded case was generated by the creation of the parameter ellipse (shown in Fig. 6). The bounds of the ellipses were modified so that the 90% confidence bounds on the expected inspection data were similar to the actual inspection data. In this way, we compensated for the lack in measured time to failure (since we have only interval data) by recognizing the information inherent in the number of items that *pass* the inspection criteria.

The simulated database can be used in various ways. Random numbers can be assigned to each item, and the items can be sorted by the random number to simulate items being processed in random order. If we sort on the creation date, we simulate processing the items in order of elapsed time after creation. It is usually advantageous to process according to age, if the parameter β is greater than 1. The overall estimated impact of such a processing strategy can be evaluated with the database. Finally we can develop a processing schedule for each material category and then process (remove from the database) the items from each material category as a function of time and determine the number of items handled in such a processing schedule that have a failed inner barrier.

Table 5. Material Category Parameters Used to Estimate Performance of the Inner Barrier

Material Category	Weibull Parameters		Calculated Inspection Results	
	Shape (α)	Scale (β)	Mean	90% Confidence Limit
Oxides	1.0	380	0.9%	4.0%
Pure Metal	1.1	210	1.8%	3.6%
Sample Returns	1.1	53.	18.6%	23.0%
Compounds and Noncombustibles	1.0	420	1.8%	3.4%
Impure Metal	1.2	280	1.6%	2.9%
Process Residues	2.3	50.	2.8%	4.5%
High-Priority Materials	1.6	48.	7.2%	10.2%

5. RISK ANALYSIS

Two characteristics distinguish this methodology for analysis of risk-based performance and processing prioritization for plutonium legacy materials from previous work in this field: (1) direct coupling between statistically treated observed data and container performance models, and (2) direct coupling between the performance model and worker risk. We considered the first characteristic in the previous section, and now develop the worker risk model in the context of the performance models established for inner barrier failure.

Two separate event trees were developed in support of the risk analysis. The two event trees were focused on the “active” risk and the “passive” risk. The differentiating factor in the event trees is the existence of an inhalation event on the “active” event tree. The single key component of this risk analysis is that the results from the statistically based random sampling work are used to estimate the probabilities for the event trees and are time-dependent.

Typically in risk analysis the conditional probabilities that determine frequencies on the event trees are derived from elicited responses from experts. This technique was avoided to create a more defensible basis for the failure frequencies. Also, it was determined that a critical feature of the risk analysis would be the time-based performance of the containers as a function of the material categories. In particular, the aging characteristics of the containers needed to be included in the risk associated with not processing certain material categories for several years. Finally, a particular subset of those out-year material categories, the Process Residues (dominated by salt materials), is known to have reactive characteristics. The performance of these materials affects the risk from changing the schedule.

5.1. EVENT TREE ASSESSMENT

The most important issue is to identify the frequencies that affect different risk outcomes for the two schedules. As a secondary issue, the frequencies that differentiate between materials types are significant because this differentiation allows prioritization of the inventory in terms of risk. Prioritization inherently lowers the difference between the two schedules.

These analyses suggest that three possible modes exist for an accident leading to worker contamination. These modes are a multiple-barrier container failure in the vault while a worker is present, a multiple container failure in the vault that is not dispersed until the container is disturbed by a worker, and an initiating-event accident while the container is in transport. Because two of these events are related to the number of containers in the vault over a period of time and not directly related to material handling, they have been termed passive failures. The other event is the active failure.

5.1.1. Event Definition

The passive event tree is shown in Fig. 7, where the frequencies shown in **BOLD** letters are derived from the sampling analysis. The value shown for the failed-fraction of Inner Container/Bagout Bag is derived from the Weibull distributions used to generate the failed fraction estimates provided previously. Since the event tree requires conditional probabilities, the frequency for failure of the outer container is on an annual basis. The number shown is derived from the results of the 100% visual inspection. The total observed failed fraction of outer containers from the 100% visual inspection is assumed to be uniformly distributed over the average age of the containers, approximately eight years. The value used for “dispersal upon disturbance” is unknown at this time. Sensitivity studies show that, in general, the induced dispersal event dominates the risk on this tree. For this reason, it is assumed for the base calculation presented in this report that 100% of all passive events are dispersed upon being disturbed. The frequency used for annual probability of container handling comes directly from the inventory processing schedules. Also, it is assumed that 10% of the containers in the vault are handled annually for surveillance purposes. Finally, a very significant event is shown on the tree, operator mitigation. Given that a container has failed through both the inner and outer containers and that vault personnel are ready to move that container (this is a disturbance), there exists a probability that vault personnel will observe the problem and take mitigating measures. For all of the base calculations in this report, it is assumed that no mitigating measures are taken.

In events where a high probability exists for operator mitigation, the top branch of the passive event tree can become significant, if we assume that a significant fraction of the containers will disperse immediately upon outer- or inner-container breach. To complete that event tree, we include the frequency that vault operators are present in a given vault room. The frequency used for base calculations is 3×10^{-3}, which represents about 30 minutes per week.

The other principal accident sequence involves the active failure. This event tree is shown in Fig. 8 and is simpler than the passive tree. The major component of the active event tree is the container drop frequency. An estimate for the container drop frequency was obtained by two different techniques.[8] Based upon information from experienced personnel, an estimated 17,000 moves have occurred since the opening of the plutonium facility. If one container drop were to have occurred out of all of those moves, the drop rate would be 6×10^{-5} per container-move. From NUREG 1278 guidelines on uncertainty, an error factor of 10 would apply. This means that the range of 6×10^{-4} would be used for the 95th percentile. Another data source is the historical information available from the Pantex Site. During the dismantlement programs at the Pantex site, drops of radioactive materials have occurred. These objects have a different character, however, than the types of containers used in the vault; the Pantex objects are heavier and more cumbersome and also must be manipulated by the operators. The drop rate data at the Pantex site lead to approximately 10^{-3} drops per move unit. To apply this to the vault situation, a performance shaping factor (PSF) must be applied. A typical PSF taken from NUREG-1278 is about 10. Thus, from the Pantex data an estimated drop rate of 10^{-4} per move-unit is expected. These results are comparable to the expected values at the plutonium facility. To provide a conservative estimate of the drop rate, the value of 6×10^{-4} was used for base calculations.

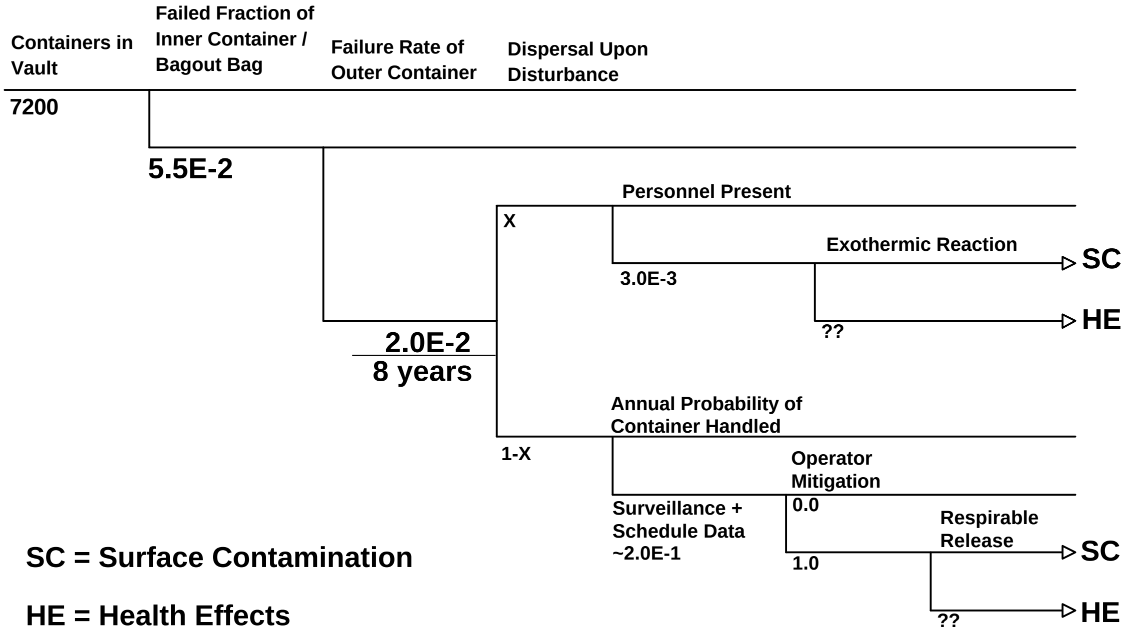

Figure 7 Event tree used for the passive-failure modes.

For the calculation of risk on the active tree, the probability of container drop is multiplied by the total number of containers handled in a given year. This number is computed in the same manner as described above; the fraction of items sampled for surveillance is added to the actual number of items processed in a given year. In this analysis, every container drop is assumed to induce a failure of either the inner or the outer container, but not both. Finally, the conditional probability that both containers fail in given a drop is computed as the probability that either the inner or outer container was failed before the drop event. This frequency is taken directly from the sampling analysis.

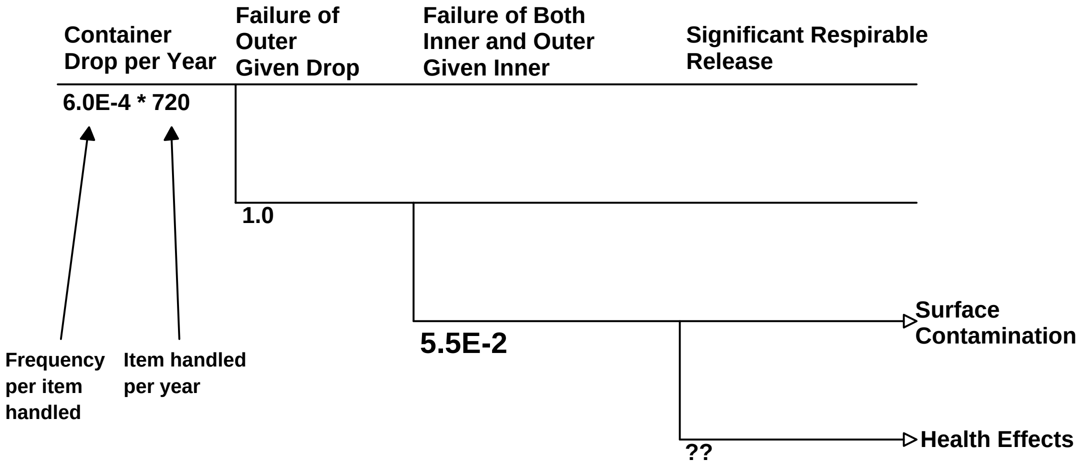

Figure 8 Event tree used for the active-failure modes with representative data.

5.1.2. Necessity for Multiple Categorization and Time-Dependency

Two key factors in this event-tree-driven risk assessment are the multiple material categorization and the time dependency. The requirements for these key factors arise because this exercise is not principally a risk assessment for the remediation program but a risk-based prioritization. Without the multiple material categories, no prioritization would be possible. However, while a large number of material categories would allow for a more highly optimized prioritization, the data requirements become excessive. Since the prioritization technique used focuses on statistical analysis as opposed to “expert opinion” wherever possible, the present number of material categories appears to achieve an appropriate balance.

Along with the multiple material categorization, modeling the time-dependent behavior of the categories from a risk perspective is also critical. This is due to the potential for highly time-dependent behavior of the material categories. The prioritization methodology must be able to evaluate the sensitivity to high-order aging effects. Without such a capability in the methodology, the appropriate impacts of delayed processing of the highly time-dependent materials, such as the High-Priority Process Residues, Compounds, and Combustibles, could not be addressed.

In Fig. 9, we estimate the number of items in the 94-1 inventory that would have failed the inner barrier had no remediation been program been instituted. We see that the number of container failures would go up almost linearly with time for most categories. This graph represents the relative gain in net risk for vault workers if the Los Alamos 94-1 program were not undertaken.

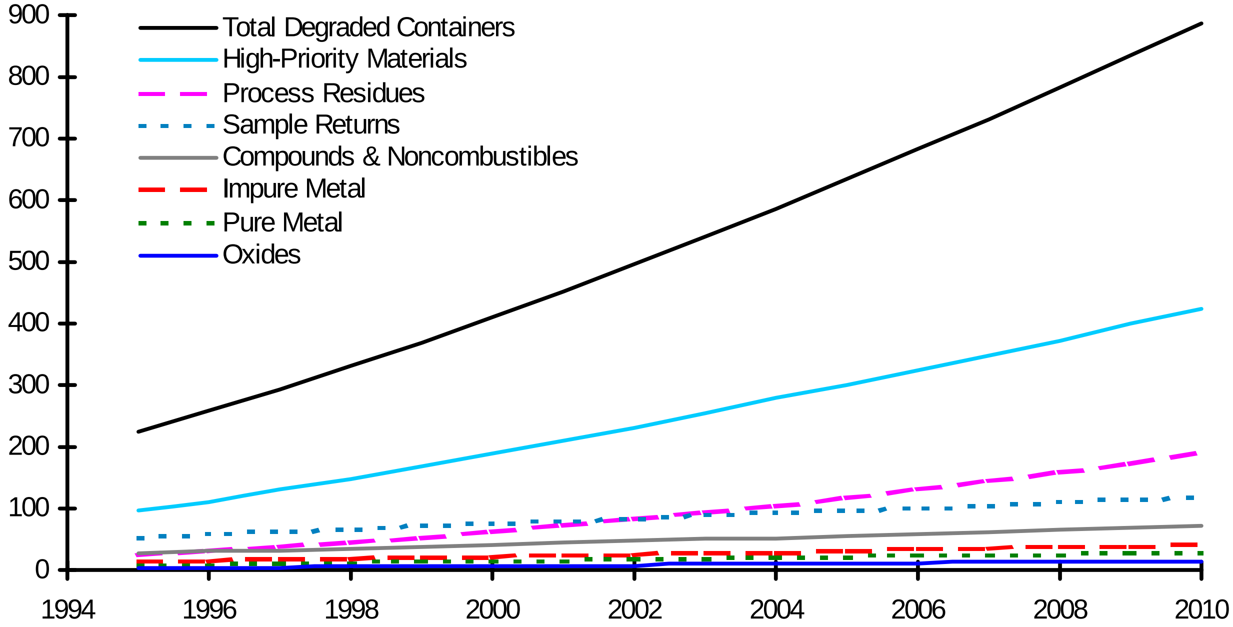

Figure 9.	Estimated number of items in Los Alamos holdings that have a failed inner barrier for the unremediated case.

### 5.2.	SCHEDULE COUPLING TO RISK MODEL

Because the interaction between integrated program risk and other factors (such as likelihood of inner barrier failure, remaining inventory, and the processing schedule) is complicated, we developed a large calculational model. Figure 10 shows how the estimated number of items in the vault with inner barrier failure decline over time as the categories are worked off. This calculation takes into account the time-dependent behavior of legacy items. In Fig. 11, we show the time-dependent probability of a risk event as defined by a vault or worker surface contamination. Note that the curves go up initially as materials are introduced to the work-off program. This is due to the increased risk associated with the number of vault operations. We show in Fig. 12 the integrated probability of risk event per unit item. This calculation serves to describe the material categories which, over the life of the program, contribute most significantly to worker risk.

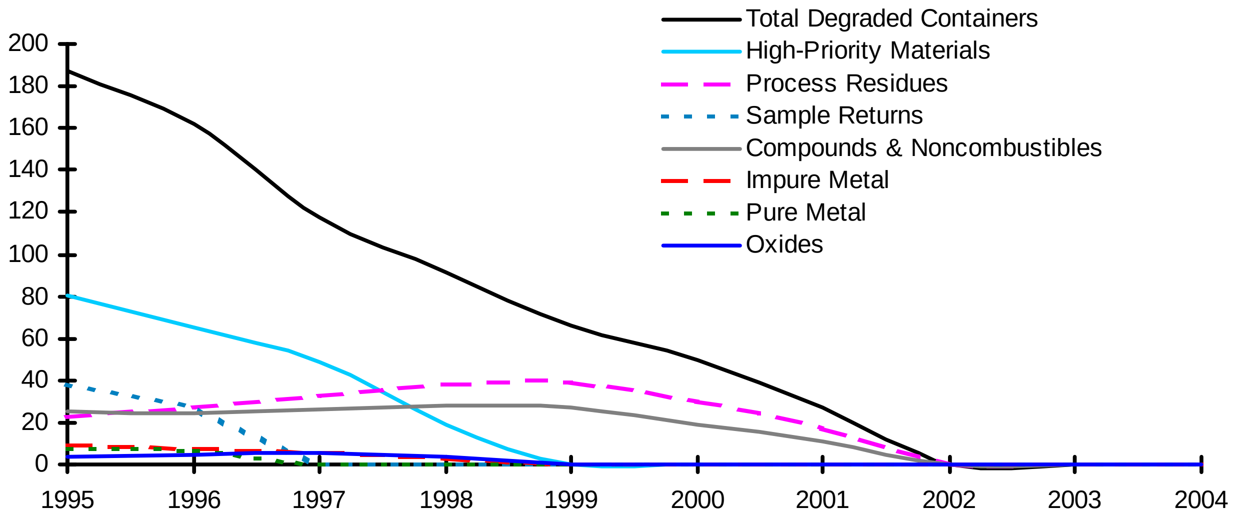

Figure 10. Estimated number of items in the Los Alamos 94-1 holdings (all legacy items) that have a failed inner barrier.

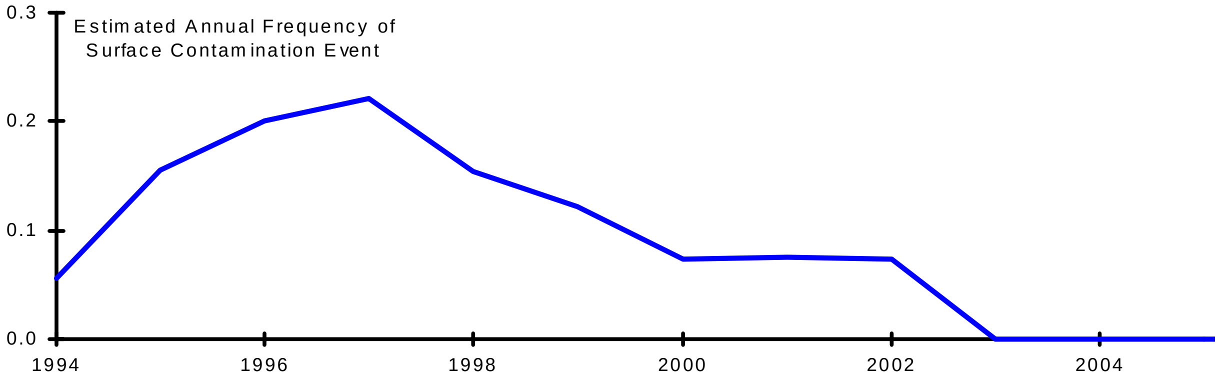

Figure 11. Estimated annual frequency of a surface contamination event for the unremediated case and for the present processing schedule.

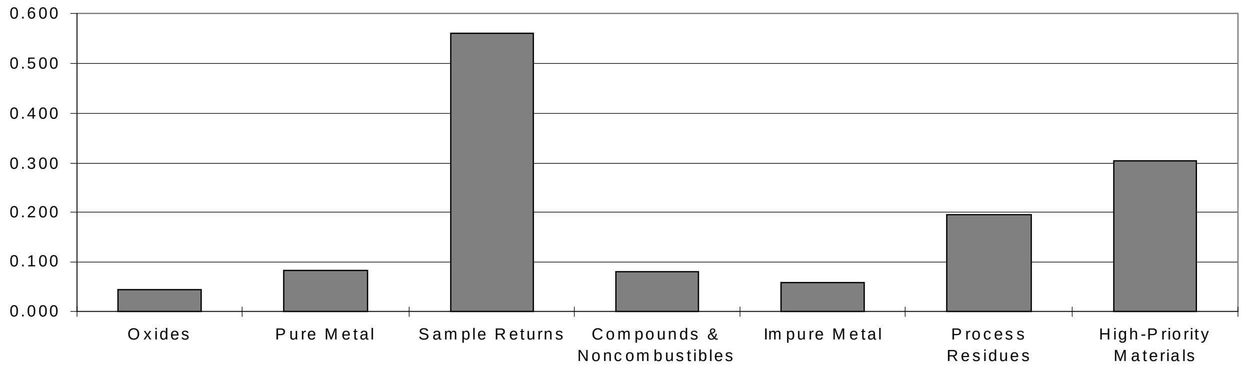

Figure 12. Integrated probability of surface contamination event per unit item for the present processing schedule.

5.3. UNCERTAINTY ANALYSIS

As we discussed in Section 4, the calculation of absolute risk is difficult and contains large uncertainties. Many other probabilities on the risk-event tree affect the overall estimation of probability of contamination event. Since we believe that these other factors are neither time-dependent nor substantially dependent on matrix material, we expect that any differences in processing strategies can be attributed to the probabilities of inner barrier failure.

Figures 13 and 14 show the results of a Monte Carlo calculation in which the present processing schedule was computed against the performance of the material categories 200 times. From these calculations, the 10% percentile, mean case (50% percentile) and 90% percentile are computed. As expected, the time-dependent risk behavior does not change very much. At the 90% case, the overall risk curve increases some, but over the entire range of time.

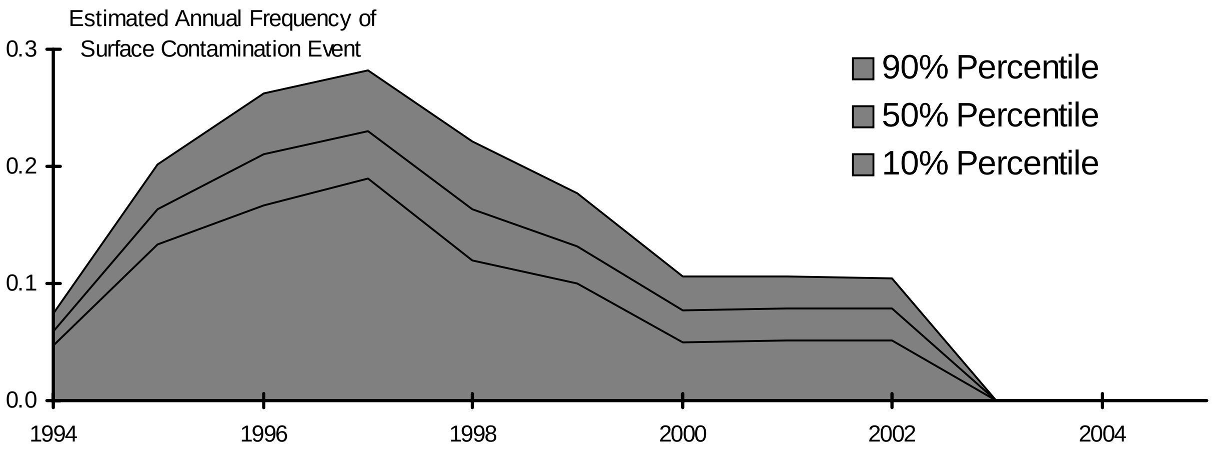

Figure 13. Monte Carlo calculation results of the annual frequency of a surface contamination event.

A more interesting result is noticed when we evaluate the normalized risk behavior. Figure 14 presents the normalized risk per unit item for the three cases evaluated in the Monte Carlo calculation. We can see the effect of the larger uncertainties on the material categories with fewer failures. In shifting from the 90% case (largest number of failures) to the 10% case (fewest number of failures) we observe that the fraction of failures for the sample returns, process residues, and high-priority materials increases. This indicates the relatively lower level of uncertainty for these categories where we have more information. Likewise, the very high level of uncertainty for the oxides category is shown by having essentially no risk in the 10% case, while a larger level of risk is computed for the 90% case.

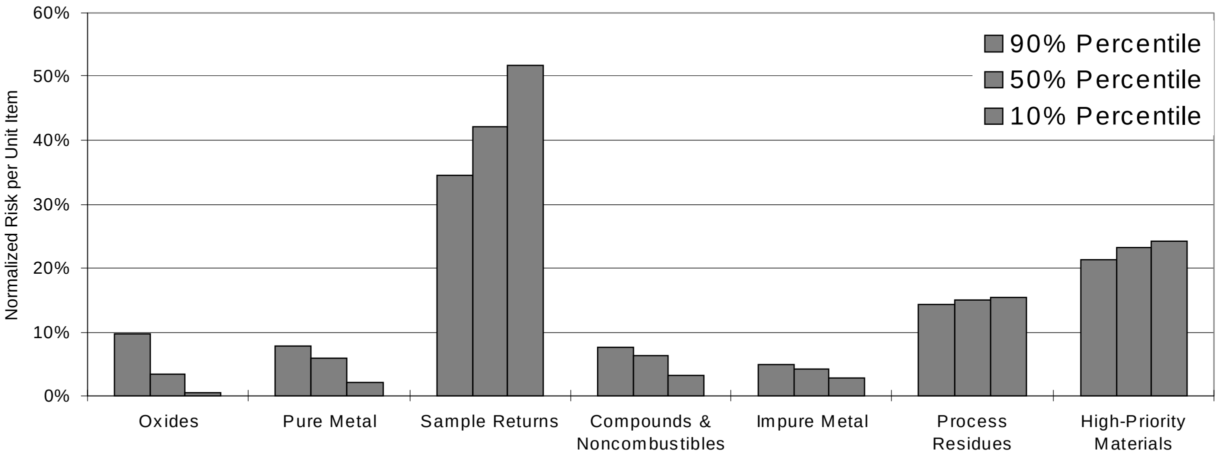

Figure 14. Monte Carlo calculation results of the normalized probability of surface contamination event per unit item for the present processing schedule.

6. RESULTS

6.1. COMPARING PROGRAMMATIC ALTERNATIVES

This methodology was developed to serve two principal tasks: provide for a risk-based prioiritization of residue processing at LANL and to provide for risk-based analysis of programmatic alternatives. Here we evaluate three specific alternatives to examine the risk impacts associated with the implementation of these alternatives: the unremediated case (No-action alternative), the unprioritized case and the 5-year delayed schedule case. In each case, we can adjust the processing schedule accordingly and compute the risk profile. These risk profiles are shown in Fig. 15. Also shown for comparison in Fig. 15 is the present schedule case (as shown in Fig. 11). Integrating the risk profile over time we compute the total program integrated risk. When we compare the program integrated risk for the processing of residues at LANL as a function of the three processing options listed above (neglecting for the moment the unremediated case), we see that the overall risk of the unprioritized case increases by about 20% as compared to the present schedule. Similarly, the 5-year delayed schedule increases overall risk by about a factor of 2. Thus, while proper prioritization of the processing program at LANL is important from a risk reduction point of view, it is more critical to be sure that the program is not delayed in implementation.

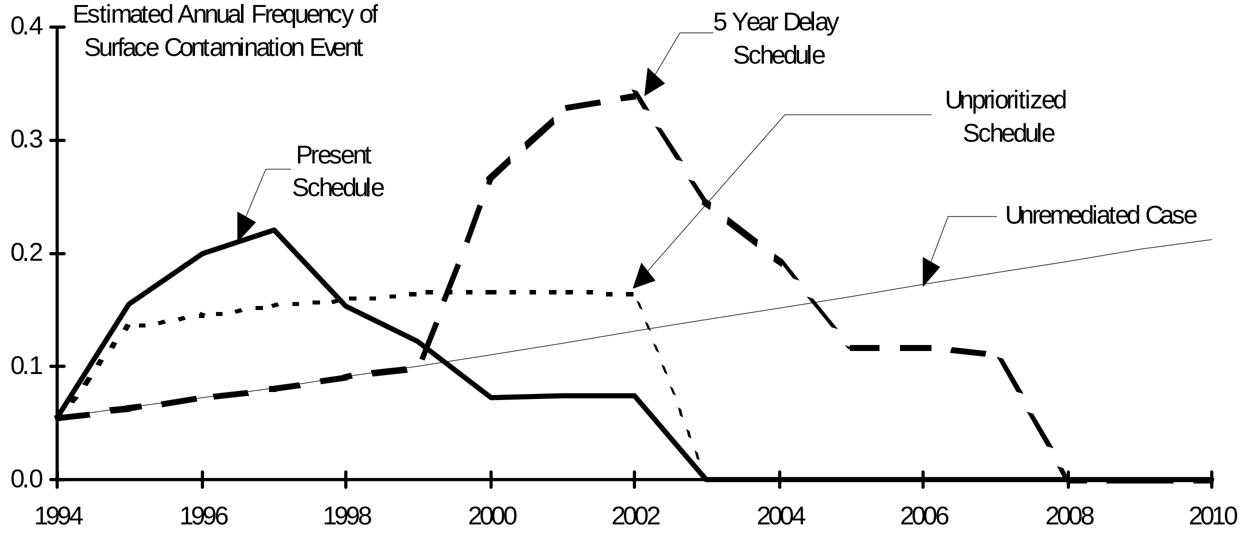

Figure 15 Time-based risk profile for three hypothetical schedules compared to the present processing schedule.

Finally, a particular critical viewpoint can be established with this risk-based program performance model. Comparing the area under the present schedule curve with the unremediated case, we find that the area under the unremediated case curve is equal to the area under the present schedule curve at about the year 2004. Another way to view these data is to compute the change in risk brought about by the present schedule using the unremediated case as a baseline. This we call comparative change in risk associated with the Los Alamos 94-1 program, and it is shown in Fig. 16. Since the unremediated case exists without a Los Alamos 94-1 program (or with complete program stagnation), we call this the “no-action alternative.” Figure 16 thus provides the *change* in risk associated with the decision to process the Los Alamos legacy inventory. Overall worker risk goes up upon implementation of the 94-1 program. This is because the nuclear materials workers are doing something more than inventory and surveillance operations required for MC&A purposes. Processing operations require workers to enter the vault much more frequently than these MC&A operations would alone. This greater level of material handling results in increased worker risk compared to the no-action alternative. After 2004, the integrated worker risk begins to shrink dramatically as a result of the 94-1 efforts.

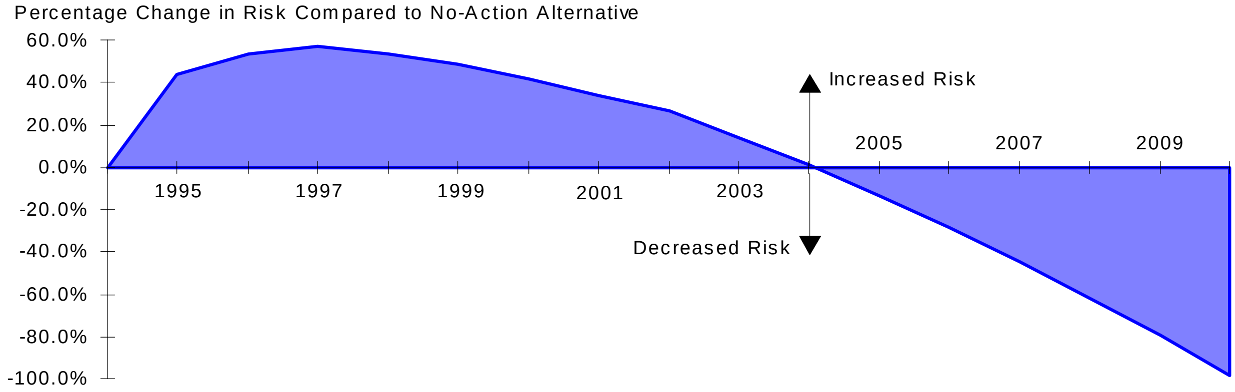

Figure 16. Percentage change in integrated worker risk, comparing the present Los Alamos 94-1 schedule to the no-action alternative.

The critical point made in Fig. 16 is that the probability of a risk event went *up* during the initial stages of the Los Alamos 94-1 program when compared to the no-action alternative. In other words, the workers increased their likelihood of a surface contamination event by engaging in the effort to remediate the plutonium legacy items. In doing so, however, they mitigated a significantly greater future hazard before it became unmanageable.

7. CONCLUSIONS

We have developed a powerful methodology for modeling the time-dependent behavior of plutonium legacy materials. The results, however, are no better than the quality of the data collected from inspections. In the past 18 months, a large body of data has been collected as part of the Los Alamos 94-1 program. For the more problematic materials, we have good confidence in understanding the fraction of items that may have a failed inner barrier.

We have been able to provide for age differences within our models, but we cannot account for other variables, such as the techniques used in packaging. The packaging techniques vary with time and location. One set of practices, associated with items returned from analytical chemistry operations, was found to have a significant impact on the stability of the residue. These sample returns were placed into a separate category, since the item source showed a far stronger indication of performance degradation than the matrix material. There are differences in contribution to risk as derived from the probability of inner barrier failure associated with different matrix materials. For example, a significant difference was found between high-priority materials and the process residues. In general, we have good confidence in our estimates of the present condition of legacy items as a function of the categories created; however, our ability to model the future time-dependent behavior is still limited for those categories with low failure probabilities because of the lack of data on item failures.

Two broad conclusions for the Los Alamos plutonium legacy materials can be drawn:

1. No evidence of strong aging effects was found. However, mild aging effects occur in several material categories. Owing to the nature of the data, it is not clear whether these effects are time-related (that is, older items were not packed as well) or are age degradation phenomena. For future risk calculations, we use the conservative assumption that these effects are degradation.

2. In the Los Alamos 94-1 remediation program, as with any remediation program, the additional handling associated with processing will raise the risk of contamination events when compared to a program without remediation. This occurs because worker risk is minimized in the near term by minimizing all handling of these materials. If, however, remediation of these materials

will be required in the future, then the sooner remediation begins and can be completed, the lower the overall program risk. While it may appear from these analyses that maximum acceleration of this and similar remediation programs would minimize the overall program risk, this is not the case. Because these activities take place within a risk management regime, proceeding at too rapid a pace can jeopardize that regime and raise the risk to unacceptable levels by effecting other nodes of the risk event tree.

Many contemporary approaches to risk management minimize the potential risk by eliminating or significantly stretching out programs. Such approaches are based on static methods for estimating risk that use information elicited from experts and are inherently incapable of forecasting risk behavior. These methods cannot dynamically adapt to data accumulated during processing, systematically compute risk reductions due to processing, or be used for risk-based determination of surveillance or sampling programs. Some have speculated that the life cycle costs for plutonium legacy remediation may become unnecessarily large as significant fractions of this material remain in an unstable state at other sites as a result of delayed implementation of remediation programs.[9] This lack of action may result from the inherent limitations of current risk management techniques, which lead to a failure to appreciate long-term risks from postponing remediation activities.

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