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Comments on a Paper Titled "The Sea Transport of Vitrified High-Level Radioactive Wastes: Unresolved Safety Issues"

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Jeremy L. Sprung, Paul E. McConnell, Paul J. Nigrey, Sieglinde Neuhauser,
Douglas J. Ammerman, Glenn F. Hohnstreiter, Richard Yoshimura

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico 87185 and Livermore, California 94550

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**Comments on a Paper Titled
“The Sea Transport of Vitrified High-Level Radioactive Wastes: Unresolved
Safety Issues”**

Jeremy L. Sprung, Paul E. McConnell, Paul J. Nigrey, Sieglinde Neuhauser,
Douglas J. Ammerman, Glenn F. Hohnstreiter, and Richard Yoshimura
Transportation Technology Program
Sandia National Laboratories
P.O. Box 5800
Albuquerque, NM 87185-0718

Abstract

The cited paper estimates the consequences that might occur should a purpose-built ship transporting Vitrified High Level Waste (VHLW) be involved in a severe collision that causes the VHLW canisters in one Type-B package to spill onto the floor of a major ocean fishing region. Release of radioactivity from VHLW glass logs, failure of elastomer cask seals, failure of VHLW canisters due to stress corrosion cracking (SCC), and the probabilities of the hypothesized accident scenario, of catastrophic cask failure, and of cask recovery from the sea are all discussed.

Acknowledgement

The authors gratefully acknowledge review of the entire report R. E. Luna and K. B. Sorenson, of Section 6.0, Release of Radionuclides from VHLW, by S. J. Glass, and of Section 9.0, Sensitization of Stainless Steel VHLW Canisters, by J. W. Braithwaite and R. Buchheit.

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1.0 Introduction.

At the request of the U. S. Department of Energy (DOE), staff of the Transportation Technology Center (TTC) of Sandia National Laboratories (SNL) have reviewed the appended paper titled "The Sea Transport of Vitrified High-Level Radioactive Wastes: Unresolved Safety Issues." The comments that follow this brief introduction refer to specific paragraphs and pages in the appended paper, which hereafter is called "**the paper**." To facilitate finding cited paragraphs, the paragraphs on each page of **the paper** have been individually numbered. The numbers are written in the left margin of **the paper** next to the paragraphs to which they correspond.

The paper poses a hypothetical ship accident scenario and then analyzes the consequences of the scenario, the release of large amounts of radionuclides from the Vitrified High-Level Waste (VHLW) and incorporation of these radionuclides into aquatic food chains. In **the paper**, all of the events that must occur, if the scenario's stated endpoint is to be reached, are not enumerated. Therefore, this commentary first attempts to fully describe the sequence of events that must occur if the stated endpoint of the **Accident Scenario** is to be reached. Next, the probability of each of the events is estimated and the estimates are used to calculate the **Scenario Probability**. Finally, the physical and chemical phenomena that **the paper** states occur as the result of the scenario are discussed. Comments about scenario phenomenology address the following six topics: (1) **Catastrophic Cask Failure**, (2) **Cask Recovery**, (3) **Release of Radionuclides from VHLW**, (4) **Individual Doses**, (5) **Elastomer Cask Seals**, and (6) **Sensitization of Stainless Steel VHLW Canisters**.

2.0 Accident Scenario.

Although **the paper** does not fully describe the ship accident scenario it hypothesizes, as the following two quotations from **the paper** indicate, **the paper** postulates that a purpose-built ship transporting two VHLW casks is struck so severely by another ship that both casks fail catastrophically, spilling their VHLW canisters into the sea.

"The most credible accident scenario that could lead to damage and loss of VHLW cargo is a collision in which the VHLW transport ship is struck in the side, where the cargo holds are most vulnerable, by the bow of another vessel. A ship with sufficient kinetic energy could penetrate the cargo holds and cause the VHLW casks to be crushed." (Page 2, Paragraph 5)

"If a damaged VHLW cask were lost at sea in shallow water, contact of the stainless steel VHLW canisters with highly corrosive seawater would begin almost immediately. Because the Type 309 stainless steel has been extensively sensitized (see below) it will undergo pitting and stress-corrosion cracking at an accelerated rate, exposing the glass underneath within a couple of months." (Page 3, Paragraph 1)

By combining this scenario with other assumptions presented in **the paper** on **Page 2, Paragraph 2 through Page 3, Paragraph 2**, the details of the accident scenario assumed in **the paper** can be developed. That development shows that the accident scenario assumed in **the paper** consists of a sequence of thirteen hypothetical events. These events are described in the following table:

Table 1. Events in the hypothetical ship accident scenario discussed in the paper

1st Hypothesis	A purpose-built double-hull ship, carrying two TN28VT shipping casks that are stowed side by side in the same hold and together are carrying 40 VHLW glass log canisters, fabricated from Type 309 stainless steel that has been sensitized by the glass pouring process,
2nd Hypothesis	is struck by a ship with a gross (loaded) tonnage ≥ 20 kton moving at a speed ≥ 20 knots,
3rd Hypothesis	while sailing over a major marine fishing region (e.g., the Grand Banks).
4th Hypothesis	The hold struck is the hold in which the VHLW shipping casks are stowed.
5th Hypothesis	The collision angle is near 90° .
6th Hypothesis	The bow of the striking ship breaches the double hull of the purpose-built ship and penetrates far enough into the hold in which the VHLW casks are stowed to apply crush forces to the casks that are large enough to cause the casks to fail catastrophically.
7th Hypothesis	Despite the fact that the stiffness of VHLW casks is substantially greater than that of the double hull of the purpose-built ship, for some reason (perhaps because the purpose-built ship has been pushed up against a second large ship) the double hull on the far side of the purpose-built ship does not fail, therefore the crush forces are not relieved, and at least one of the two VHLW casks fails catastrophically.
8th Hypothesis	Crushing of the cask causes all of the glass logs in the VHLW canisters in the cask that fails catastrophically to fracture extensively which increases the surface area of the glass fragments by about a factor of ten above the surface area the logs had when originally cast.
9th Hypothesis	Because the purpose-built ship is cut in two, both casks fall into the ocean and during settling to the ocean floor the 20 VHLW canisters in the failed cask spill from that cask onto the ocean floor.
10th Hypothesis	Although the casks are massive and thus easy to detect and the VHLW canisters are highly radioactive, poorly shielded, and thus also easy to detect, the casks, canisters, and/or VHLW fragments are not found and recovered in less than 60 days.
11th Hypothesis	During this 60-day period, corrosion of the sensitized steel canisters that spilled onto the ocean floor now causes those canisters to fail catastrophically in a short time (about 30 days).
12th Hypothesis	After the exposed canisters crumble away, exposure of the VHLW glass log fragments to ocean waters leads to leaching of the glass during the next 30 days at a rate of about 2×10^{-6} g $\text{cm}^{-2} \text{d}^{-1}$.
13th Hypothesis	This leach rate releases enough radioactivity into the marine fishing region so that entry of the radionuclides leached from the fragmented VHLW glass logs into the marine food chain causes individuals, who consume as a part of their normal diets very large amounts of sea foods harvested from this region, to receive doses during the year that follows the collision that exceed the recommended limits for members of the public and also the recommended limits for occupational exposures.

The following section estimates the probability of this ship accident scenario.

3.0 Scenario Probability.

Although values are not available for many of the events in the scenario set forth in Table 1, an estimate of the overall probability of the scenario can be made. Thus, let

P_{acc} = the probability per nautical mile sailed of a collision at sea

N_{mi} = the number of nautical miles sailed in order to traverse the fishing region in which the accident is postulated to occur

P_{hold} = the probability that the hold containing the two VHLW casks is struck

P_{pen} = the probability that the double hull is breached and the hold penetrated far enough to subject the VHLW casks to crush forces

P_{cat} = the probability that the crush forces are not relieved by pushing the casks through the double hull on the far side of the struck hold; thus the probability of catastrophic cask failure

P_{can} = the probability that the canisters spill out of the cask onto the ocean floor

P_{cor} = the probability that the temperature, salinity, and pH of the ocean water where each canister settles is such that extensive stress-corrosion cracking occurs in about 30 days

P_{rec} = the probability that recovery efforts fail to locate and recover most of the VHLW canisters in ≤ 30 days

Reasonable estimates for these probabilities and for the number of miles sailed in the fishing region are:

$P_{acc} = 2 \times 10^{-7}$ per nautical mile sailed in coastal water [1].

$N_{mi} = 150$ miles, the width of the continental shelf [2].

$P_{hold} = 0.125$.

Pacific Nuclear Transport Limited purpose-built ships have 5 cargo holds and additional length (the bow and the crew and machinery spaces) equal to 3 additional holds; therefore $P_{hold} = 1/8 = 0.125$.

$P_{pen} \leq 0.1$ [3].

$P_{cat} = 2 \times 10^{-7} = (2 \times 10^{-7} \text{ per nautical mile sailed})(1.0 \text{ nautical mile})$

The chance that, during a collision at sea, the purpose-built ship will be pushed up against another object large enough to prevent the VHLW casks from being pushed through the double-hull on the far side of the hold is extremely small; essentially the chance that a collision with another ship or possibly an oil platform will immediately follow the first collision.

$P_{can} = 0.25$.

The chance that the cask is oriented downward during its fall to the ocean floor is 0.5 and the chance that crushing of the cask prevents the canisters from spilling out of the cask is estimated to be 0.5.

$$P_{\text{cor}} = 10^{-2}.$$

The chance that ocean conditions promote extensive stress corrosion cracking and thus catastrophic canister failure within 30 days; and

$$P_{\text{rec}} = 10^{-2}.$$

The recovery actions that followed the crash of TWA flight 800 show that a major recovery effort on the North American continental shelf can be mounted in days; for the Pu shipments from Japan to Europe, the casks were equipped with transponders to facilitate locating them should they be lost at sea; even if the VHLW casks are not equipped with transponders, as the canisters are highly radioactive, they will be easy to locate and because they are not bulky or heavy, recovery using a submersible with robotic arms should not be difficult.

Substituting these values now leads to a value of approximately 10^{-18} for P_{scenario} , the probability of this scenario. Thus, the probability that this scenario might actually occur is extremely small, so small that the scenario is hardly credible.

$$P_{\text{scenario}} = P_{\text{acc}} N_{\text{mi}} P_{\text{hold}} P_{\text{pen}} P_{\text{cat}} P_{\text{can}} P_{\text{cor}} P_{\text{rec}} \\ = (2 \times 10^{-7})(150)(0.125)(0.1)(2 \times 10^{-7})(0.25)(10^{-2})(10^{-2}) = 2 \times 10^{-18}$$

4.0 Catastrophic Cask Damage.

As the quotes from **Page 2, Paragraph 5** and **Page 3, Paragraph 1** of the paper indicate, the paper assumes that the hypothesized ship collision will cause at least one VHLW cask to fail catastrophically, spilling all of its VHLW canisters onto the ocean floor. Catastrophic failure with loss from the cask of all of its canisters must be assumed to occur, if the canisters are to be fully exposed to flowing seawater. Full exposure of the canisters to flowing (not quiescent) seawater must be assumed, if the canisters are to fail catastrophically within a short time (i.e., within a couple of months, as is postulated in the paper) as a result of stress-corrosion cracking. Rapid catastrophic canister failure must be assumed, if the glass fragments in the canisters are to be subjected to leaching by freely flowing seawater for at least 30 days before the fragments can be recovered.

During most accidents involving a purpose-built ship carrying a VHLW cask, the cask would not be severely damaged; that is, catastrophic failure would not occur because the structure of VHLW casks is more robust than both the structure of the purpose-built ship transporting the cask and the bow structure of almost all possible striking ships. Because of the robust design of the casks, the most probable result of a collision that subjected the cask to crush forces would be that the cask would be pushed across the hold in which it is stowed until it is pushed up against the double-hull structure on the side of the ship opposite the collision. The amount of force that could be imparted to the cask by such an accident is limited by the stiffness of the double hull of the purpose-built ship. Since the double hull is less stiff than the cask, unless there is some additional structure on the far side of the ship's double hull (e.g., unless the purpose-built ship is

pushed up against some structure robust enough and massive enough to act like an unyielding surface), any crush forces applied to the cask will be relieved by collapse of the double hull and not by yielding of cask structures.

Determining the magnitude of the force applied to a cask trapped between the penetrating bow of a striking ship and the double hull of the struck ship, the purpose-built ship, would require a detailed analysis of the specific design of the purpose-built ship and, to a lesser extent, of the damage to the struck ship and to the bow of the striking ship caused by the collision. Analyses now being performed at Sandia that neglect damage to the bow of the striking ship [4] show that, for one specific single-hull structure, the forces experienced by the cask would be about the same order of magnitude as the forces experienced by Type B packages during impacts onto unyielding surfaces. The double hull of purpose-built ships (e.g., the fleet of purpose-built ships operated by Pacific Nuclear Transport Limited) probably would not produce a force acting on the package that is significantly larger than those seen for the single-hull ship used in this Sandia analysis because the space between the two hulls is sufficiently large that penetration of the first hull would occur before the second hull is contacted. Even for forces much larger than those seen during qualification of Type B packages, it is unlikely that the package would be severely damaged, much less fail catastrophically. A recent test program subjected a simulated cask similar to a VHLW cask to very severe impacts with no failure of the containment boundary. The maximum "rigid body" accelerations seen in the test series were about 1200 Gs. This is about 7 times higher than the acceleration expected for this type of package during a regulatory impact test [5].

Furthermore, if some mechanism could be envisioned that would subject a Type B cask (e.g., the TN28VT cask considered in **the paper**) to a force large enough to cause it to fail catastrophically (e.g., crush forces sufficient to severely warp the seat of the cask lid causing failure of most of the lid bolts and probably loss of the lid), then inward deformations of the cask walls would be so large that the VHLW canisters would be trapped inside the cask by the crushing of the cask walls and basket structures about the canisters [6]. Then, if the cask were to be lost into the sea, as the canisters would still be trapped within the cask, the cask would substantially limit the flow of ocean water past the canisters.

Page 2, Paragraph 5: The accident scenario implicitly assumed in **the paper** supposes that the transport ship is struck by another ship with sufficient kinetic energy to penetrate the cargo hold and crush the VHLW cask. **The paper** cites no basis for this assumption. In fact, results of finite element analyses reported by McConnell et al. [7] indicate that the compressive crush forces experienced by a cask, when caught between the bow of a striking ship and the far wall of a struck ship, are smaller than the compressive forces experienced by the cask when first impacted by the bow of a striking ship, which in turn are lower than the impact forces to which a cask is subjected during the regulatory 9-meter drop test, because the impact velocity of the striking ship is lower than the regulatory impact velocity and the bow of the striking ship is less stiff than the unyielding impact surface used in the regulatory test.

5.0 Cask Recovery (salvage).

Page 2, Paragraph 4 and Page 5, Paragraph 1: The paper quotes (see reference 3 in the paper) part of one sentence from a report issued by the Organization for Economic Cooperation and Development (OECD) without presenting the following sentence and succeeding discussion of purpose-built ships and cask recovery plans that largely negate the conclusion of the quoted sentence. The quoted sentence is

This shows that coastal transportation accidents are therefore unacceptable.

The omitted sentence that immediately follows the quoted sentence is

In the next section, it will be shown, however, that it is possible to design a transportation system and to plan recovery actions in case of an accident, so that the probability of occurrence of these doses and the resulting risk can be made extremely small.

In the OECD report, the section that follows this sentence describes the design of a double-hull purpose-built ship and the plans for recovery of any cask lost at sea from such a ship, and concludes that the ship design and the likelihood of recovery of any cask lost at sea from such a ship mean that transport of VHLW waste in purpose-built ships is safe.

Page 5, Paragraph 2: The paper does not consider the extensive discussion of salvage operations presented in the OECD report that it cites on **Page 2, Paragraph 4 and Page 5, Paragraph 1**. The OECD report assumes salvage within one year. Even for packages not equipped with transponders, the probabilities of locating casks lost at sea on the continental shelf and of finding the contents of the casks intact are both estimated in the OECD report to have values of 1.00. The OECD report gives a lower probability of 0.50 for a ruptured penetrator (which would contain a VHLW canister), and gives probabilities of recovery for casks, canisters, and damaged canisters of 0.99, 0.99, and 0.90, respectively (p. 163). Therefore, the concerns presented in the paper about the feasibility of recovery are unfounded.

Page 5, Paragraph 3: Because the paper assumes that canister failure and extensive leaching of radionuclides from the fractured glass logs will all occur within a few months, it concludes that recovery activities would also have to be completed within a few months since radiation doses to salvage crews would become unacceptably large once large quantities of nuclides had been released from the glass. But the TWA 800 crash shows that large complicated recovery operations can be initiated in a few days. Therefore, because large metal objects (for example, a VHLW cask) and highly radioactive materials (the glass log inside of a VHLW canister) are easy to detect, detection and recovery within a few months of the cask and any canisters spilled that from the cask as a result of the accident is likely. Moreover, as canister corrosion rates and glass erosion and radionuclide leaching rates are all likely to be significantly slower than is assumed in the paper (see discussions of canister corrosion and erosion and leaching of glass presented below), recovery most likely does not need to be completed as rapidly as is assumed in the paper.

6.0 Release of Radionuclides from VHLW.

Page 3, Paragraph 2: The paper states that a number of studies have identified Cs-137, Am-241, and Cm-244 as the largest contributors to doses from accidental loss of a VHLW cask at sea. As no references are provided for this statement, it could not be verified. In addition, the VHLW inventory presented on **Page 1, Paragraph 1 of the paper** for 1021C glass contains more Cs-134 (121 TBq) and Sr-90 (3220 TBq) than Am-241 (101 TBq) or Cm-244 (95.6 TBq), which suggests that, for the 1021C glass in which the VHLW is to be shipped, Am-241 and Cm-244 would be minor rather than major contributors to population dose.

Page 3, Paragraph 3: The paper uses dynamic leach-rate data from the study of Vernaz and Godon [8] for the leaching of Am-241 from R7T7 glass to estimate the leach rate of Am-241 from 1021C glass. But Vernaz and Godon measured a dynamic leach rate for Am-241 from SON 61 glass that is a factor of 3 smaller than the dynamic leach rate of Am-241 from R7T7 glass. Because leach rates depend strongly on the composition of the glass from which the leaching occurs, use of the higher result for R7T7 glass rather than the lower result for SON 61 glass, or perhaps the average of the two results, should have been justified in **the paper** (i.e., 1021C glass should have been shown to be more like R7T7 glass than SON 61 glass, if the SON 61 results were to be neglected, as is done in **the paper**). More importantly, because the leach rates reported in the study of Vernaz and Godon are all normalized leach rates (i.e., the leach rate of the radionuclide divided by the mass concentration of the radionuclide in the glass sample), the leach rate value for Cs used in **the paper** is 300 times larger than the actual measured leach rate for Cs because the normalizing factor for Cs = $g_{Cs}/g_{Glass} = 1/300$ [9].

Page 3, Paragraph 4: The Paper states that "the presence of salt generally accelerates the dissolution of glass in water," and cites (see reference 4 of **the paper**) the following sentence as the basis for the statement:

The same is true for the influence of salt, which can lead to significant acceleration of dissolution.

but omits the sentence that immediately follows this sentence:

Thus, it has been found with a soda-lime silicate glass that at 80° C after 100 hrs the weight loss in 1-molar solutions of LiCl, NaCl, or KCl amounts to 102, 189, and 431 $\mu\text{g}/\text{cm}^2$, respectively, while it amounts to only 7 $\mu\text{g}/\text{cm}^2$ in pure water.

And, more importantly, neglects this sentence from page 332 of the same book:

With borosilicate glasses, according to the kind of glass, either higher or lower values were found.

where the "higher or lower values" are the solubility of the glass in water that contains dissolved salts. Thus, **the paper** neglects the strong dependence of glass erosion and leaching rates on

glass composition and, despite this dependence on composition, applies results for a soda-lime silicate glass to a borosilicate glass, while also neglecting results for other non-borosilicate glasses that conflict with the result cited for the soda-lime silicate glass. To be specific, the experiments of Barkatt et al. show [10] that, for a synthetic microtektite glass with a silica content close to that of VHLW glasses, "the dissolution rates in seawater are lower by more than a factor of 30 than the rate observed in deionized water,..." **The paper** also neglects leaching data for VHLW glass exposed to brines which show that, for the temperature range 25 to 350°C, leach rates for brines are nearly an order of magnitude lower than leach rates in distilled water [11]. Thus, **the paper** cites one result that is favorable to its argument and neglects many other results that indicate that erosion and leaching of VHLW glasses may also be retarded by the presence of dissolved salts in the leach water.

In the same paragraph, **the paper** also claims that the static leaching results of Vernaz and Godon for R7T7 glass indicate that "the presence of salt enhances the release of glass constituents such as boron and plutonium (therefore indicating a general increase in the glass alteration rate) by a factor of approximately 4." **The paper** does this without noting that another study that it cites (the OECD study cited in Reference 3 of **the paper**) [12] reaches a different conclusion, specifically "that in seawater the leaching rate is the same [as for distilled water] or, at a maximum, increases by a factor of 2." More importantly, **the paper** is misinterpreting the results of Vernaz and Godon. Specifically, the factor of 4 increase was obtained by Vernaz and Godon during an experiment where the leaching solution was water saturated with NaCl that also contained 2 g of solid NaCl. Thus, the increased leach rate is primarily caused by adsorption of the leached material onto the solid salt and not by the use of brine as the leach solution. In fact, other experiments performed by Vernaz and his coworkers [9] have shown that that for leaching in brine "there is no increase in leach rate, as there is no increase in sea water."

Page 3, Paragraph 5: The **paper** relies on the raw data given in Figure 2 of the Vernaz and Godon paper to generate the Cs-137 leach rate ($2.2 \times 10^{-6} \text{ g cm}^{-2} \text{ d}^{-1}$) that it applies to the first 30 days that follow the hypothetical catastrophic failure of the canisters, which it assumes will be caused by stress-corrosion cracking. **The paper** claims to have obtained this leach-rate value by time-integration of the first 30 days of data in this figure. In order to perform this integration, some mathematical distribution must have been assumed. Since neither a distribution nor its parameter values are specified in **the paper**, it is impossible to determine whether the initial leach rate derived in **the paper** is appropriate.

Furthermore, **the paper** segregates the first 30 days of data from later data without citing any justification for doing so. As the discussion in the next paragraph shows, this segregation is, in fact, not justified.

Macedo et al. [13] note with regard to glass leach tests, that "data taken during the first few days are of little value." Macedo et al. also restate the well-known observation that "during the initial period of exposure of glasses to water, the dissolution rate is usually high, then falls off rapidly as a function of time." They explain that this effect has two distinct causes. Glass dissolution rates are elevated initially because of "the rapid dissolution of fines, surface irregularities, and siliceous impurities adsorbed on the sample during processing or preparation for the test." Thus,

initial dissolution rates depend strongly on sample preparation. Consequently, they are not true leach rates. Initial dissolution rates are also rapidly decreased by the formation of a protective layer on the surface of the glass. Specifically, when immersed in water, a protective, hydrated layer, that inhibits erosion and leaching, rapidly forms on the surface of glasses and other vitreous materials, and even forms on glasses in damp atmospheres. Seven days was the time period for protective-layer formation noted by Jantzen et al. [14] for a polyphasic ceramic material; and a layer approximately 10 microns thick already had formed by day 10 (earliest data point recorded) on vapor-hydrated glass [15]. Because protective layer formation on glass is rapid, infiltration of seawater into the VHLW canisters through cracks formed in the canister by canister crush or by stress-corrosion cracking would cause the glass in the canister to hydrate while the glass was still protected by the canister walls before those walls could fail catastrophically due to the prolonged effects of stress-corrosion cracking. Thus, for the scenario postulated in **the paper**, a protective-layer would have formed on all VHLW glass log surfaces long before the time when the glass first became fully exposed to free-flowing seawater. Therefore, the use in **the paper** of a leach-rate that typifies the first 30 days of VHLW leaching is improper, because regardless of whether it takes two months (the estimate in **the paper**) or many decades (more likely) for the canisters to corrode sufficiently to fully expose the fragmented VHLW logs to free flowing seawater, a protective, leach-resistant layer will already have formed.

Thus, to summarize, the use of an accelerated leach rate for the first 30 days of exposure of glass fragments to freely flowing seawater is inappropriate as the leach rate is most likely not accelerated during this period, principally because a protective layer will have formed on the surface of the glass long before catastrophic canister failure occurs (if indeed it does occur).

Protective-Layer Formation. Inhibition of glass erosion by formation of a protective layer is strongly supported by studies of ancient glasses, volcanic glasses, and microtektites.

Studies of Ancient Glasses. Archaeological data have become available in the last few years that can be used to put an upper bound on the leach rates of ancient glasses submerged in the ocean. Two wreck sites -- the 3000-year-old Ulu Burun wreck and the 1000-year-old Serce Liman wreck, both located off the coast of Turkey -- have been carefully excavated under the direction of George Bass and Cemal Pulak of the Institute for Nautical Archaeology (INA) [16-20].

The Ulu Burun site is an irregular, rocky slope averaging about 150 ft in depth that frequently experiences a strong current. The ship had carried among other items a cargo of cobalt-blue glass ingots (i.e., raw product meant to be melted down and used by the eventual buyer to make finished objects). Many of these ingots (5-6 inches in diameter and several inches high) remained essentially intact beneath a hydrolyzed layer several millimeters thick, although others were extensively hydrolyzed. Some of the latter were found within rocky pockets on the seabed where organic material from the wreck (e.g., ship timbers and wickerwork) had also collected, and it is possible that these ingots had been exposed to acidic decomposition products that accelerated hydrolysis; others apparently had extremely frizzled, irregular, bubbly or cracked surfaces to begin with, which increased the depth of the

hydrolyzed layer. The fact that they were produced as a raw material and turned out of the molds with little care for the condition of the surface is believed to be the reason the glass surface was sometimes in this condition. This glass was manufactured at only a single location in the ancient world; and well-preserved, nonsubmerged specimens have been excavated by archaeologists, so the original size of the ingots could be readily established. The glass beneath the hydrolysis layer, incidentally, was still quite blue (i.e., still retained all of its cobalt).

Cobalt also was found in the chemical analysis of earthenware amphorae recovered from the site, and is known to not be present in contemporaneous examples from terrestrial sites, which suggests that redeposition of some of the dissolved cobalt probably occurred. This phenomenon is likely to be a general one and would tie up some fraction of the ions dissolved out of any glass, which indicates that dispersal models such as MARINRAD are conservative in this respect.

The glass recovered from the Serce Liman wreck consists of thousands of fragments of broken glass from finished objects with smooth surfaces. One of the objects, a large bottle with walls originally about 4 mm thick, has been completely reassembled by the INA. The fragments had been packed into baskets for use as ballast. Strong currents were also noted at this wreck site. The hydrolysis layer for the Serce Liman glass averaged about 1 mm in thickness, although greater and lesser degrees of hydrolysis were noted (e.g., there are a few holes in the reassembled bottle; conversely, some glass fragments are still sharp enough to cut one's hand [21]). Where a group of fragments could be identified as having a common origin (as in the fully reassembled bottle), the original thickness could be readily estimated from those fragments that retained their sharp edges, since such a condition is presumptive of a very low degree of hydrolysis.

Dr. Robert H. Brill of the Corning Museum of Glass has performed chemical analyses on glass objects from both of these wrecks. He believes that as a rule of thumb, ancient soda-lime glasses submerged in seawater hydrate at a long-term rate of about 1 mm per 1000 yrs, but emphasizes that this rate is very sensitive to glass composition [22]. He pointed out, for example, that the Ulu Burun glass, with a hydrolysis layer averaging around 4 mm in thickness, was slightly higher in alkali and, therefore, slightly less durable than the Serce Liman glass. Although all ancient glasses are soda-lime glasses, not the Pyrex-like borosilicate glasses used to make VHLW, the fact that so much of the glass remained after being exposed to seawater for one and three millennia, respectively, supports the OECD's suggestion that long-term leach rates are much lower than extrapolation from short-term data would suggest. This reduction in long-term leach rate can be attributed to the formation of a protective layer on the surface of the glass. It also can be argued that, since borosilicate glasses are generally far more durable than ancient glasses and since protective layers also form on these glasses, they would hydrolyze at rates no greater than and probably lower than those observed for ancient glasses.

Studies of Volcanic Glass and Microtektites. Dr. Brill also notes that the leach rate of volcanic obsidian is one-thousandth of that of ancient glasses [22], a result consistent with the findings of A. Barkatt, et al. [10] who report that microtektites "exposed to aqueous environments for 720,000 years" have corroded to depths "not more than 20 μm ."

Page 3, Paragraphs 6: In this paragraph, **the paper** further increases the leach rate it applies to the first 30 days of exposure of glass fragments to freely flowing seawater by assuming that the surface area of the glass logs carried in the canisters is increased by a factor of ten as a result of the crushing of the canisters during the accident. Specifically, **the paper** takes the surface area of one canister ($19,000 \text{ cm}^2$) and multiplies that value by ten, claiming that this accounts for the "extensive fracturing" of the glass that would be caused by the hypothesized accident. **The paper** does this even though a Pacific Northwest Laboratory study shows [23] that VHLW glass logs in canisters subjected to a 9-meter drop onto an unyielding surface experience little fragmentation. Multiplication of the accelerated initial leach rate of $2.2 \times 10^{-6} \text{ g cm}^{-2} \text{ d}^{-1}$, that the paper assumes applies to the first 30 days of glass fragment exposure to freely flowing seawater, by $190,000 \text{ cm}^2$, the canister surface area increased by a factor of ten, then leads to the leach rate of 0.42 g d^{-1} assumed in **the paper** to apply to the first 30 days after catastrophic canister failure.

7.0 Individual Doses.

Page 4, Paragraph 1 and Table 1: Table 1 of **the paper** presents values for "committed effective doses to individuals resulting from radionuclide releases in shallow waters." All of the estimates of peak individual doses presented in **the paper** are calculated using the values of committed effective doses taken from the third column in this table labeled "OECD (1988) best estimate." However, despite careful review of the cited OECD report, we were unable to find or derive these "OECD best estimate values."

Page 4, Paragraph 5: Even if the values labeled OECD values in Table 1 are correct, **the paper's** use of these values to predict short-term population doses is not appropriate because the OECD report explicitly states that significant population doses do not occur until ten years after the accident. Further, **the paper** presents no justification for its application of these values to a much earlier time period. In the highly improbable event that canisters were to be lost in the manner assumed in **the paper**, the ten years that must elapse before population doses become significant means that recovery of the cask, canisters, and/or log fragments during the first few months after the accident (as the discussion of **Cask Recovery** above suggests is likely) will ensure that the population doses that might be caused by such an event would be significantly smaller than the doses estimated in **the paper**, as very little radioactivity will have escaped from the glass before recovery takes place.

Page 4, Paragraph 6: The objections presented in the discussion of peak doses (see **Page 4, Paragraph 5** above) also apply to collective doses. Accordingly, collective dose would not begin to be accumulated until many years after the accident, again ensuring that considerable time would be available during which to recover the cask, canisters, and/or glass log fragments lost in the ocean waters above the continental shelf.

8.0 Elastomer Cask Seals.

Page 4, Paragraph 3: The statement in **the paper** that the damaged cask analysis performed by the Central Research Institute of the Electric Power Industry of Japan (CRIEPI) "assumes the shipping cask remains intact except for the O-ring seal" is false. Tsumune et al. [24] of CRIEPI, in their PATRAM '95 paper, specifically consider release of radionuclides into seawater through loss of sealing function of the packaging, seawater coming into the packaging, loss of containment of the canisters, and exposure of the vitrified waste container to seawater. They consider the environmental impact for a near shore sinking and for a sinking at sea. From their studies, the dose equivalent that the public would see was calculated for the near shore and deep sea cases to be $\sim 6 \times 10^{-4}$ mSv/yr after 2 months submergence, and $\sim 5 \times 10^{-9}$ mSv/yr after submergence for 45 years. **The paper** also states that CRIEPI failed to take into account ocean currents. This latter point is again incorrect, as the CRIEPI researchers specifically considered advective or ocean current velocities in their calculation. These omissions by **the paper** reflect a selective reporting of technical details.

Page 5, Paragraph 5: **The paper** states that elastomers fail after several hours when exposed to temperatures in the range of 250 - 300°C but does not mention which type of elastomer it assumes is failing in that temperature range. Type B casks, such as the TN29VT cask, are usually equipped with elastomer seals that are made from materials which have good resistance to heat, for example, fluorocarbons (e.g., Viton) or silicones. For Viton and silicone elastomers, recommended temperature ranges for normal service (where normal service lifetimes are defined to be 1000 hours) and exposure times for service at temperatures above the recommended normal operating range are presented in Figures A3-5 and A3-6 in the Parker O-Ring Handbook [25]. Figure A3-5 shows that the high end temperatures for the recommended temperature range for normal (1000 hr) service for Viton and silicone elastomers are 204 and 232°C respectively, and that both compounds will survive short-term exposures to temperatures of about 260°C. Figure A3-6 shows that both compounds should survive one-hour exposures to temperatures of about 300°C. Because these temperature limits are conservative, actual failure should require longer exposure times than those given in the Parker handbook.

Results presented at PATRAM '95 indicate that, when held for three hours at temperatures as high as 288°C, Viton and silicone O-rings do not fail [26]. During these experiments, seal leakage was checked upon reaching the test temperature, after holding the seal at the test temperature for two hours, after cooling to ambient temperature, and finally after cooling to minus 40°C. If a helium gas leakage rate in excess of 1.0×10^{-7} cm³/sec was measured during any of these tests, then the O-ring was judged to have failed.

Because water has a larger molecular weight and a much larger viscosity than helium gas, the permeation of water through elastomer seals will be much lower than that of helium. Therefore, Viton and silicone seals would be expected to be leaktight to seawater even if they had previously encountered temperatures of 250 to 300°C for several hours.

In the same paragraph, **the paper** also states that elastomers are damaged when exposed to high radiation fields. But the sealing locations in transportation packages are specifically designed to provide a significant degree of shielding for the seals to the radiation emitted by the package contents. Thus, the discussion in **the paper** of degradation of elastomers due to radiation damage fails to account for the ability of the packages to attenuate the radiation levels which means that the elastomer seals are subjected to radiation levels significantly less than the high radiation levels that exist at the surface of the VHLW glass logs.

Page 5, Paragraph 6: In this paragraph, **the paper** asserts that during normal transport the cask seal comes dangerously close to its 250°C failure threshold and cites results of Yamakawa et al. [27] as the basis for the conclusion. First, as **the paper** correctly notes, the results of Yamakawa et al. show that VHLW cask seal temperatures under normal operating conditions are about 148°C. But the high end temperatures for the recommended normal operating range for Viton and silicone elastomers are 204 and 232°C, respectively (see **Page 5, Paragraph 5** above). As service lifetimes at recommended normal operating temperatures are at least 1000 hours, seal failure is hardly imminent under normal cask transport conditions.

Page 6, Paragraph 1: **The paper** states that, "when a prototype VHLW transport cask was tested by Japanese authorities, the seal temperature reached 178°C following exposure to Type B thermal [fire] conditions, an increase of 30°C," and then asserts that exposure to such a fire would cause seal failure "after approximately 2.5 hours." The paper then asserts that radiation damage to seals "may lower the temperature threshold or reduce the time to failure at elevated temperature." As the discussion of **Page 5, Paragraph 5** above shows, Viton and silicone elastomer seals are expected to survive for at least 1000 hours at 178°C and for several hours if heated to about 300°C, the temperature **the paper** assumes the seals reach if heated by a regulatory fire for 2.5 hours. Moreover, as failure means leakage of helium and not water, should the accident proposed in **the paper** involve exposure of the cask to a fire for several hours before the casks fall into the ocean, leakage rates for water into the cask still would be of no concern. The discussion of **Page 5, Paragraph 5** above also shows that cask design limits radiation damage to seals. Thus, exposure of seals to radiation is not expected to synergistically increase seal failure times during fires. Finally, contrary to the assertion of **the paper**, ship fire probabilities have been extensively studied, and most ship fires are of short duration or, if of long duration, are smoldering fires that spread slowly from one location to another and do not reach temperatures in any location that are high enough to threaten cask seals.

9.0 Sensitization of Stainless Steel VHLW Canisters.

Page 3, Paragraph 1: In **the paper** seawater is said to be highly corrosive. This is misleading since seawater has a pH of 8.0 ± 0.5 and thus is only slightly basic [28].

Page 6, Paragraph 2: No evidence is presented in **the paper** that the thermal cycle to which the VHLW canisters are exposed due to the pouring of glass causes a phase transformation or sensitization of SUH 309 austenitic stainless steel. Although sensitization may be possible,

quantitative information regarding the thermal (time/temperature) history of the steel is necessary to estimate its extent.

Page 6, Paragraph 3: Steel designated “SUH 309” apparently refers to a grade of heat-resisting austenitic stainless steel specified in Japan Institute of Standards (JIS) Standard Number G4311 [29,30] for which there is no exact international equivalent [31]. SUH 309 has a general chemical composition and a chromium and nickel content similar to that specified for austenitic Cr-Ni heat resisting steel under Unified Number S30900 (which includes, for example, ASTM A473 Type 309 stainless steel) in the Unified Numbering System for Metals and Alloys [32]. The specified carbon level for SUH 309 is $\leq 0.20\%$. **The paper** states on **Page 6, Paragraph 3**, however, that the steel has 0.15 wt % carbon and, in **End Note 17**, that COGEMA reports “that the carbon content of the ‘Type 309’ stainless steel that it is using has been lowered to 0.08 %.” Thus, the exact composition of the canister stainless steel is unclear. But precise specification for the canister stainless steel is important (1) to judge the consistency of the sources cited in **the paper**, (2) because the carbon content of the steel directly influences the degree of sensitization, and (3) because some grades of “309” stainless steel contain niobium and possibly tantalum to stabilize carbides, which mitigates sensitization. Thus, because the exact composition of the canister stainless steel (and of the thermal transients to which the canister is subjected) is not presented in **the paper**, **the paper’s** assessment of canister sensitization can only be approximate.

Page 6, Paragraph 3: **The paper** reports that in an experiment a “stainless steel with a carbon content of 0.08 % sensitized completely after only 30 minutes.” This may be true, but one must consider the temperature at which the experiment was conducted. Temperatures near the high end of the “400-850 °C” sensitization range greatly reduce the time required for sensitization and vice versa. At the low end of the sensitization range, an exposure time of two hours would not cause an 18Cr-9Ni stainless steel with a carbon content of 0.08 % to become sensitized [33, p. 377].

Page 6, Paragraph 4: **The paper** states without providing a reference that “COGEMA data shows that the VHLW canister temperature remains within the sensitization range for about 7 hours.” Without detailed data about the thermal history (and carbon content) of the VHLW canister, degree of sensitization can not be assessed. Moreover, data on the filling of canisters with glass at the U.S. DOE Savannah River Site indicates that during the glass pouring step, canister temperatures are not elevated long enough for significant sensitization to occur. Specifically, Plodinec and Harbour report [34] that during the filling of Defense Waste Processing Facility canisters at the Savannah River Site “[t]he maximum temperature of the canister wall for a continuously filled canister was 425 to 500°C. Generally, the steel, at a particular level, will be above 400°C for approximately two hours. Overall the time of exposure [of a canister] to this [canister filling] heat treatment is only a few hours and is at a temperature below that required to get significant sensitization. This relatively short heat treatment further reduces the potential formation of embrittling phases and chromium carbide precipitation. Therefore, this heat treatment due to glass-filling will not significantly enhance corrosion of the canister”

The statement in **the paper** that the COGEMA and BNFL VHLW canisters experience a thermal excursion during filling that causes significant sensitization requires verification; it seems likely that the thermal history of the VHLW canisters is similar to the relatively benign temperature excursion of the DWPF canisters. If the assertion on **Page 6, Paragraph 4** that "the canister temperature remains within the sensitization range for about 7 hours" can be substantiated, then sensitization of the VHLW canisters is likely.

Page 6, Paragraph 6 through Page 7, Paragraph 1: Although not convincingly proven by **the paper**, if it is assumed that the stainless steel in the VHLW canisters has become sensitized, then the integrity of the canisters will be determined by localized intergranular corrosion or stress corrosion cracking (SCC) enhanced by the sensitization and not by general corrosion because, as **the paper** recognizes, the average rate of corrosion over the entire canister surface will be extremely slow. Factors necessary for stress corrosion cracking are stress, existence of chloride ions, aqueous solution with dissolved oxygen, and a sufficiently elevated temperature. In a sub-sea surface environment these factors are, in turn, affected by submersion depth (temperature) and whether the canisters are embedded in sediments (pH). For a submerged VHLW canister all of the factors needed to promote stress corrosion cracking, except for a sufficiently elevated temperature, will be present. However, the low temperatures that characterize ocean environments will strongly retard the rate of localized corrosion of a submerged, exposed VHLW canister, as substantially elevated temperatures are required for stress corrosion cracking to proceed at a significant rate.

The paper suggests an ambient seawater temperature of 10°C, a temperature so low that corrosion kinetics should be quite slow. Water temperatures of $\leq 5^{\circ}\text{C}$ to 20°C are typical of seawater at all latitudes, depending upon ocean depth [35]. Heating of the canister due to decay of radionuclides in the VHLW glass will cause the surface temperature of the canister to be somewhat higher than the temperature of the seawater in which it is immersed. A steady-state calculation for a stainless steel TN28VT canister immersed in 10°C water indicates that the surface temperature of the VHLW canister will be only about 3 to 5°C higher than the temperature of the seawater in which it is immersed. "[C]hloride SCC does not occur in nonsensitized austenitic stainless steels at temperatures below about 60°C in near-neutral chloride solutions [such as seawater, pH ~ 8]. Under severe [stress or loading] conditions this minimum temperature can be lowered to 50°C and to temperatures approaching ambient for sensitized [stainless steel]" [33, Figure 63].

A. J. Sedricks [36] states that "[t]emperature is by far the most important variable in determining whether chloride cracking will occur [For] austenitic stainless steels, chloride cracking is not a hazard at ambient temperature [T]his applies also to welded stainless steels in ambient marine atmosphere, [although] attack can occur in severely sensitized materials" The potential for cracking of a stainless steel as a function of pH, temperature, and chloride content is depicted by a figure in the texts of Sedricks [36] and also of Jones [37]. For neutral pH and a chloride content of 10^4 ppm (seawater contains $\sim 1.9 \times 10^4$ ppm Cl^- ions), no cracking is observed until temperatures exceed the boiling temperature. Pitting may be observed at temperatures as low as 40°C , but pitting per se would not cause any structural damage to a canister, nor would

pitting result in the leaching of the canistered waste [38]. Higher temperatures and more acidic conditions (pH values less than that of slightly alkaline seawater) may, however, enhance the propensity for cracking.

Sensitization can be detrimental to chloride SCC resistance. However, papers [39-48] that discuss intergranular stress corrosion cracking in chloride and/or acidic environments indicate that such cracking only occurs in elevated temperature environments ($> 25^{\circ}\text{C}$).

No evidence of localized corrosion of sensitized stainless steel at normal ocean temperatures (i.e., less than 25°C) has been found. Failure of VHLW canisters exposed to normal ocean temperatures would not be expected to occur due to localized corrosion within a period of "a few months' time." Assertions within **the paper** that the VHLW canisters will fail when exposed to seawater, e.g., "... will virtually guarantee that the canisters will fail" (see **Page 7, Paragraph 1**) and "... stress corrosion cracking at an accelerated rate [will] expos[e] the glass underneath within a couple of months" (see **Page 3, Paragraph 1**), are speculative and are neither convincing nor verifiable. Even if cracking due to corrosion should occur, it is unlikely that the integrity of the canisters would be impaired to the extent that they would fall apart on the ocean floor over periods of time as short (a few months) as those assumed by **the paper**.

Even if the stainless steel became sensitized and chloride-assisted SCC occurred, crack propagation rates in the stainless steel canister walls would be low. For fully sensitized Type 304 stainless steel, immersed in a concentrated 22% NaCl solution at $\text{pH} \approx 7$ at temperatures from approximately $20\text{-}50^{\circ}\text{C}$, the SCC crack growth rate of a highly stressed test specimen is only about 5×10^{-8} to 10^{-7} mm/s [49]. For a canister wall thickness of 5 mm, this suggests that, if SCC should initiate, through-wall crack propagation would require 5×10^7 to 10^8 seconds, that is, about 19 to 38 months.

Page 6, Paragraph 7: The assertion in **the paper** that "reporters . . . were warned not to touch [VHLW canisters] to prevent exposure of the steel to potentially corrosive salts in their sweat" greatly exaggerates corrosion rates by equating by inference the negligible corrosive effect of a "touch" to the degradation and failure of a canister exposed for a long period of time to a seawater environment.

Page 8, End Note 18: The comment that "the stainless steel VHLW canisters experience tensile stresses in storage at or above their yield strengths" requires documentation.

10.0 Other Comments.

Page 1, Paragraph 4: **The paper's** assertion that "the industry" has not provided a satisfactory response is incorrect. In fact, the International Atomic Energy Agency (IAEA) in 1994 initiated a Coordinated Research Program (CRP) titled "Accident Severity at Sea during the Transport of Radioactive Material" specifically to evaluate a number of the issues raised by **the paper**. The prime objective of this CRP was to insure the validity of the principle of providing safety

"designed in" to packages. Five IAEA member states are currently contributing to this program. The CRP includes probability studies, fire studies, impact studies, and assessment of the radiological consequences of radioactive material transport at sea. The PATRAM '95 conference held in December of 1995 had an entire session devoted to sea transport. As one of the papers presented at this conference is cited in **the paper**, it is curious that **the paper** neglects the other papers presented at this conference by other researchers in the U.K., France, Germany, Japan and the U.S. in this area, especially when they reach conclusions that contradict those presented in **the paper**.

11.0 Summary

Review of the appended paper suggests

- (1) that the scenario analyzed is so improbable that it is of very little or no concern;
- (2) that should the scenario be initiated, its endpoint, catastrophic failure of a VHLW cask followed by the spilling of all of the VHLW canisters in the cask onto the ocean floor, is most unlikely to be reached;
- (3) that even if, against all odds, the scenario endpoint were somehow to be reached, the temperature, pH, and salinity of seawater above the continental shelf are such that the rates of stress corrosion cracking of VHLW canisters, of erosion of VHLW glass logs, and of leaching of radionuclides from eroded logs would be quite slow;
- (4) that given these slow rates of canister corrosion, glass log erosion, and radionuclide leaching from eroded logs, the leached radionuclides will enter aquatic food chains very slowly over a lengthy period of time;
- (5) that this slow entry of radionuclides into aquatic food chains (at least two orders of magnitude slower than predicted in **the paper**) will ensure that the population and individual doses delivered through those chains are small compared to normal background exposures; and
- (6) that all of these processes (corrosion of canisters, erosion of glass logs, and leaching of radionuclides from eroded glass logs) will almost certainly be prevented from proceeding to an extent of concern by recovery (salvage) of the cask, canisters, logs, and/or log fragments before more than a few (2 or 3) months have passed.

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Appendix

The Sea Transport of Vitriified High-Level Radioactive Wastes: Unresolved Safety Issues Preliminary Report for Comment Edwin S. Lyman, PhD

**NUCLEAR CONTROL
INSTITUTE**

Washington, D.C.

Preliminary Report for Comment

The Sea Transport of Vitrified High-Level Radioactive Wastes: Unresolved Safety Issues

**Edwin S. Lyman, PhD
Scientific Director
Nuclear Control Institute
December 1996**

Introduction

1 In February of 1995, the ship *Pacific Pintail* set sail from Cherbourg, France for the port of Mutsu-Ogawara in northern Japan, carrying a shipment of 28 canisters of vitrified (glassified) high-level radioactive wastes (VHLW). This shipment of extremely hazardous material, the first of dozens being planned to return VHLW generated during the overseas reprocessing of Japanese-origin spent fuel, ignited considerable controversy at many points along its sea route. In the face of united opposition by Caribbean states, the ship avoided the Panama Canal and instead sailed around Cape Horn, the tip of South America, where it antagonized Brazil, Argentina and Chile.

2 When the *Pintail* reached Japan in late April of 1995, the governor of Aomori Prefecture initially refused to permit the ship to dock, employing a resolution which granted him the authority to refuse entry into port of any vessel if he felt that the assurance of safety and the provision of information to be inadequate. The standoff was resolved a day later, after he had extracted a promise from the Science and Technology Agency (STA) that it would not authorize a final disposal site for HLW in Aomori without the governor's consent.

3 In my view, the worldwide unpopularity of the *Pintail* and its cargo was entirely justified. In late 1994, when I was a postdoctoral research associate at the Center for Energy and Environmental Studies at Princeton University, I was commissioned by three environmental and nuclear non-proliferation advocacy organizations (Greenpeace International, Nuclear Control Institute and Citizens' Nuclear Information Center) to examine safety issues associated with the upcoming sea transport of VHLW. Initially, I was skeptical that these shipments posed significant environmental risks. However, as I began to analyze the technical and regulatory basis for the safety of shipping VHLW by sea as it was being practiced, I found it seriously deficient in many respects. In particular, I found that the degree of uncertainty surrounding the potential performance of each component of the shipping system was so large as to make assurances of safety practically worthless.

4 The report of my findings ¹ was released in December 1994. The public response of the nuclear industry to it was to attack my qualifications, my integrity and my independence.² However, the industry was unable to refute the principal technical points in my report, and in retrospect it has stood up quite well. Also, additional information has since come to light that raises new questions.

5 Now, nearly two years later, the industry is about to launch another shipment of VHLW. This shipment which will consist of 40 VHLW canisters, is a bridge between the initial, pilot shipment of 28 canisters and subsequent full-scale shipments of 150 canisters each. This shipment will proceed despite the fact that the industry has not provided satisfactory responses to the serious safety concerns raised in my 1994 report. These issues, which include the use in the TN 28 VT VHLW shipping cask of elastomer seals which can lose their function in a fire of moderate severity, and the use by COGEMA and BNFL of a type of stainless steel for packaging the VHLW which loses its corrosion resistance during VHLW processing, are reviewed below.

Details of the Second VHLW Shipment

1 Japan has released a description of the 40 VHLW canisters which will be transported in the second sea shipment, including an inventory of radionuclides. The composition of the VHLW canister labeled "1021C" is typical (although below average for the shipment); some of the more significant radionuclides are:

Americium-241 (Am-241): 101 Terabequerels (TBq)

Curium-244 (Cm-244): 95.6 TBq

Strontium-90 (Sr-90): 3220 TBq

Cesium-134 (Cs-134): 121 TBq

Cesium-137 (Cs-137): 5000 TBq

Total alpha emitters: 210 TBq

Total beta-gamma emitters: 18,000 TBq

Total heat generation: 1.5 kilowatts (kW)

2 The second shipment will consist of two casks carrying 20 VHLW canisters each, while the first shipment consisted of one cask carrying 28 VHLW canisters. One explanation for this is that the average heat generation of the second shipment will be 1.61 kW per canister, which is slightly higher than the average for the first shipment, which according to measurements made by Japanese authorities after delivery, was 1.54 kW/canister.

The Consequences of a Loss of VHLW Cargo at Sea

3 One type of accident that was not considered in detail in the 1994 report was one in which the ship and its VHLW cargo is lost at sea. A closer examination of this accident scenario indicates that it can potentially result in severe health consequences for humans and for marine biota. However, the large uncertainties inherent in calculations of this type, especially for accidents in which the cargo is lost in coastal waters, result in extremely wide ranges for predictions of potential consequences. This is clear from a survey of studies which attempt to evaluate the consequences of the loss of a VHLW cask at sea, and obtain results which differ by as much as six orders of magnitude (a factor of 1,000,000). The predictions of doses to the public, which range from very low to highly significant, are sensitive to numerous assumptions, such as the location of the accident, the condition of the VHLW packages, the leach rate of the glass in seawater, the local current patterns, food consumption, the extent to which radionuclides would be sorbed on sediments and the likelihood of salvage.

4 However, it is important to note that in the worst case, the loss of a damaged VHLW cargo in coastal waters can cause levels of chronic exposure to the public far in excess of those permitted by international standards. This fact was pointed out in an 1987 Organization for Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA) study of the feasibility of disposing of VHLW under the ocean floor, an undertaking that would require by necessity the large-scale transport of VHLW by sea. The analysis contained in this study led to the conclusion that "coastal [VHLW] transportation accidents are ... unacceptable."³

5 The most credible accident scenario that could lead to damage and loss of VHLW cargo is a collision in which the VHLW transport ship is struck in the side, where the cargo holds are most vulnerable, by the bow of another vessel. A ship with sufficient kinetic energy could penetrate the cargo holds and cause the VHLW casks to be crushed. The OECD study recognized that because the consequences of this accident would violate its own safety criteria, it was necessary to design a transport system in which the probability of occurrence of such an accident would be "extremely small." To meet this goal, the study proposed the use of purpose-built ships designed so that their holds could not be penetrated by ships of any displacement (mass) travelling at a speed of 24 knots (12.3 m/s) or below.

6 The Pacific Nuclear Transport Limited (PNTL) ships that transport VHLW today are supposedly designed to be resistant to collisions. This type of ship is designed so that if it is struck by a ship of 24,000 tonnes displacement travelling at 15 knots then the cargo area will not be penetrated. However, the protection is afforded by this design criterion is limited, since there are ships in service which have much greater displacements and travel at much greater speeds. Some ships travel at speeds up to 25-30 knots, and displacements of tankers can exceed 100,000 tonnes. A ship with a 50,000 tonne displacement traveling at 30 knots would have a kinetic energy eight times greater than the ship modeled in the design basis collision with a PNTL vessel.

1 If a damaged VHLW cask were lost at sea in shallow waters, contact of the stainless steel VHLW canisters with highly corrosive seawater would begin almost immediately. Because the Type 309 stainless steel has been extensively sensitized (see below) it will undergo pitting and stress-corrosion cracking at an accelerated rate, exposing the glass underneath within a couple of months.

2 A number of studies have identified the radionuclides cesium-137 (Cs-137), americium-241 (Am-241) and curium-244 (Cm-244) as those responsible for the largest contributions to both individual and collective doses for a loss of VHLW in coastal waters. In order to calculate these doses, one must know two pieces of information: first, what are the rates of release of these radionuclides under the appropriate conditions, and second, how these radionuclides are transported from the release point through the marine food chain and finally to humans.

a) Release of radionuclides from VHLW

3 The leach rates of radionuclides from VHLW in contact with deionized water are fairly well-characterized. According to a recent French study,⁴ the room temperature dynamic leach rate (e.g. water replenished daily) of Cs-137 from R7T7 reference glass is 2×10^{-7} g/(cm²-d) (grams per square centimeter of surface area per day), equivalent to 6.4×10^{-7} terabequerels (TBq) per square centimeter per day. For Am-241, the leach rate for R7T7 glass doped with 1.1 GBq/g Am-241 is 3×10^{-7} g/(cm²-d);⁵ for the composition 1021C, which contains about 0.22 GBq/g Am-241, the corresponding leach rate would be 6.7×10^{-8} g/(cm²-d), or 8.5×10^{-9} TBq/(cm²-d). Similarly, the leach rate of Cm-244 from 1021C is found to be 1.9×10^{-8} g/(cm²-d), or 5.8×10^{-8} TBq/(cm²-d).

4 Less information is available for VHLW leach rates in contact with highly saline seawater. However, the presence of salt generally accelerates the dissolution of glass in water.⁶ For static leaching of R7T7 glass at 90°C, the presence of salt enhances the release of glass constituents such as boron and plutonium (therefore indicating a general increase in the glass alteration rate) by a factor of approximately 4.

5 Previous studies of the loss of a VHLW cask have assumed these or similar values. One flaw of this assumption is that it does not take into account that these values are steady-state average release rates. However, when water first comes in contact with VHLW, the initial leach rates are higher by a factor of around 100 (prior to the formation of a less permeable surface layer) and then decrease steadily over the course of about a month until they reach the steady-state values.⁷ Integration of the exponential transient leach rate versus time curve shows that the average Cs-137 release rate for the first month is 2.2×10^{-6} g/(cm²-d), eleven times higher than the steady-state value. The same increase is true for actinide release rates. This means that as much Cs-137 would be released in the first month following contact of the VHLW surface with water as would be released in the following eleven months. The initial leaching behavior is important for determining how soon salvage of the cargo would have to occur to avoid significant release of radiation into the environment.

6 Using the above room-temperature leach rates in pure water (room temperature is valid, because even though the ambient water temperature on the continental shelf is around 10°C, the VHLW itself is a significant heat source and will raise the temperature of the glass-water interface by several degrees at a minimum), a VHLW geometric canister surface area of 19,000 cm² and an effective surface area ten times greater (because the VHLW is extensively fractured), the radionuclide release rates per VHLW canister for the first few years after the accident are approximately:

Cs-137: 1.4 TBq/d (first 30 days); 0.12 TBq/d (subsequently)

Am-241: 0.018 TBq/d (first 30 days); 0.0016 TBq/d (subsequently)

Cm-244: 0.12 TBq/d (first 30 days); 0.011 TBq/d (subsequently)

7 Leach rates in seawater may be as much as four times greater. Over the course of a fifty year period, radioactive decay will attenuate the rate of emission of Cs-137 (half-life = 30 y) and Cm-244 (half-life = 18.1 y) by factors of two to three. The Am-241 rate (half-life = 432 y) will decrease much more slowly.

b) Conversion of releases to exposures

8 Calculation of the radiation exposures to the public that would result from the release of radionuclides from a damaged VHLW cask lost in coastal waters is a complicated and uncertain undertaking. One issue is the complex and site-specific nature of the ocean current flows in these regions. A review of four studies shows that there can be large variations in estimates of the

contributions of different radionuclides and pathways to final exposures.

i) Individual doses

The following table illustrates this disparity by comparing the coefficients which relate unit releases of the above three radionuclides to the 50-year committed effective doses (CED) to individuals that can result. These coefficients are usually not explicitly given and have to be extracted from other data provided in the reports; they are therefore approximate. It is important to note that it has been shown that the models used to compute doses are linear with respect to the radionuclide leach rate; therefore, these results can be used for the R7T7 leach rates given above. It should also be noted that the dose computations in all of these studies (with the exception of Nielsen [1996]) were originally based on ICRP 30 data. ICRP 30 has since been replaced with ICRP 68, in which the coefficients expressing dose per unit intake of ingested actinides have been reduced (e.g. Am-241 by a factor of 3 and Cm-244 by a factor of 2.5); the data in Table I has been adjusted accordingly.

TABLE I

**Committed Effective Doses to Individuals Resulting from
Radionuclide Releases in Shallow Waters (mSv/TBq)**

Radionuclide	Klett (1986) ⁸ (average)	OECD (1988) "best estimate"	CRIEPI (1995)	Nielsen (1996) ⁹
Cs-137	0.002	0.002	1×10^{-8}	0.02
Am-241	0.001	0.44	2.3×10^{-7}	0.4
Cm-244	0.0004	0.24	8×10^{-7}	not given

On the low end of the scale are the results of a study done in 1995 by the Central Research Institute of [the] Electric Power Industry of Japan (CRIEPI).¹⁰ The largest disparities in the table exist between the CRIEPI results and the others, although there is a significant difference between the OECD and Klett (1986) results for the actinides (which is odd because they are both based on the same model).

The CRIEPI results differ so dramatically from the others because they make a number of assumptions which are unreasonable. CRIEPI's "damaged cask" assumes that the shipping cask remains intact except for the O-ring seal, so that water can only enter the cask through the narrow gap between the cask lid and body. CRIEPI also assumes that the only mechanism for release of radionuclides from the cask is by natural convection and diffusion. These assumptions lead to a very slow leak rate from the cask. However, there are appreciable currents near the sea bottom in coastal waters (up to 2 cm/s), and depending on the orientation of the cask, these could facilitate flushing of the cask contents. The only way to explain the anomalously low dose rates obtained by CRIEPI is to assume that even if the O-ring fails, the cask will inhibit the release of radionuclides by a factor of as much as one million. The level of conservatism inherent in such an assumption is questionable, to say the least.

Excluding the CRIEPI results, the studies seem to agree to within a factor of 10 on the individual CEDs resulting from Cs-137 releases.

If the OECD values in Table I are used, one finds the following results for the peak individual CEDs, summing over all three radionuclides: 2.3 mSv/canister after the first year, 1.3 mSv/canister-year for subsequent years. For 20 canisters, this corresponds to 46 mSv after the first year, and 26 mSv/y for the first few years afterward. Alpha-emitting radionuclides (Am-241 and Cm-244) are responsible for 97% of the total CED. *These are huge dose rates, exceeding not only the ICRP 60 limit for exposure of the general public (1 mSv/y), but also the limit for those occupationally exposed (20 mSv/y). Enhancement of the leach rate by the presence of salt could increase these doses by as much as a factor of four.* An average individual receiving the above (decay-corrected) radiation exposure for a fifty-year period would have a 7% risk of contracting a fatal cancer as a result of this exposure.¹¹ In the U.S., this would correspond to an increase in lifetime risk of cancer mortality of more than 25%.

ii) Collective doses

Of the four studies reviewed above, only two of them, OECD (1988) and Nielsen (1996), calculated collective dose commitments resulting from radionuclide releases in shallow waters. It is not as

straightforward to adjust these results for the 1021C source term. However, it is possible for a few radionuclides. According to the OECD results, the 50-year integrated collective dose resulting from an initial Am-241 leach rate of 0.038 TBq/yr is 1.84 person-Sv (again adjusting for ICRP 68), and for Cs-137, it is 21.8 person-Sv for an initial leach rate of 1.65 TBq/yr. Scaling these results for 20 canisters of R7T7 VHLW gives a 50-year collective dose of 566 person-Sv from Am-241 and 1.2×10^4 person-Sv from release of Cs-137. This collective dose from these two radionuclides alone would cause around 650 fatal cancers in the 50-year period.

It is clear from these results why the OECD recognized that the loss of a VHLW cask in shallow waters would indeed have "unacceptable consequences." Frankly, it is unknown whether the current shipping system operates so that the risk of such an accident would be acceptably low.

PNTL assumes that if a VHLW cask were lost in shallow waters, it would be immediately salvaged. However, they have provided no evidence to the public that they would be capable of carrying out such a hazardous operation, especially if the cask were damaged. In fact, at a March 1996 meeting at the International Maritime Organization, the question of salvaging potentially damaged, highly radioactive cargoes was discussed. It became clear in the course of the discussion that issues specific to this cargo that may affect the feasibility of salvage operations have not been carefully thought out.

As shown above, salvage of a damaged VHLW cargo would have to be carried out within a few months of the accident to prevent a substantial release of contamination. Once that has occurred, salvage operations would become immensely difficult, posing great health risks for the salvage crew. Raising the entire ship would probably be the only tenable option, but such an operation would likely facilitate the spread of contamination to surface waters. Until the industry provides a credible salvage plan that addresses these contingencies, the public cannot give much credence to industry assurances that salvage will be carried out.

Safety Issues Previously Raised that Remain Unresolved

In the 1994 report, a number of issues with implications for the safety of VHLW sea transport were raised. Industry has never provided to the public a satisfactory resolution of them.

a) Elastomer cask seals

The TN 28 VT VHLW transport cask utilizes a lid seal made of an elastomeric (rubber-like) material. The lid seals play an essential role in preventing the escape of radioactive gases and fine particulates from the cask should an accident occur. However, elastomeric materials have poor heat resistance and will fail after exposure for a couple of hours to temperatures in the vicinity of 250°-300°C; above this range, they will fail in under one hour. Furthermore, elastomers are damaged by exposure to high radiation fields. For these reasons, elastomer seals do not appear to be the best choice for casks transporting heat-generating, highly gamma-emitting materials like spent fuel or VHLW, especially when compared to costlier metallic seals, which offer superior heat and radiation resistance.¹²

In 1994, my report pointed out that the thermal power of a loaded TN 28 VT cask was so great that the temperature of the cask seals during normal transport was dangerously close to the 250°C failure threshold; I estimated that it would lie in the range 120°-170°C. In December 1995, a paper by CRIEPI on thermal studies of the TN 28 VT cask was presented at an international meeting which found the seal temperature to be 148°C and thus verified my estimate.¹³

(One should also note that the CRIEPI report found that the centerline temperature of the VHLW canisters in the fully loaded cask was 390°C. The actual temperature of the 1995 transport was higher (probably greater than 400°C) because the average heat loading of the VHLW canisters shipped was 1.54 kW, with some as high as 1.65 kW, greater than the 1.46 kW assumed in the Japanese test. When my report pointed out in 1994 that the cask was designed so that the centerline temperature could be as high as 510°C, it was ridiculed. Calling the report "dubious science," Gavin Carter of BNFL told the Journal of Commerce that the temperature "...will be 200 to 250 degrees Centigrade."¹⁴ Of course, my report never stated that the temperature was 510°C, only that such high temperatures were permissible. In any event, we now know that the actual temperature was closer to 510°C than to 250°C.)

Transport casks with elastomer seals are able to be qualified as IAEA Type B packages because the heat input generated by the Type B thermal test is low enough so that the seal temperature remains below the failure threshold, even with a high initial seal temperature (provided the cover protecting the seal remains intact following the impact tests). But the current regulations do not require the cask designer to determine the conditions which *would* cause the seal to fail and to ensure that a large safety margin is present.

For instance, when a prototype VHLW transport cask was tested by Japanese authorities, the seal temperature reached 178°C following exposure to Type B thermal conditions, an increase of 300°C. While this result was judged to provide a "sufficient safety margin" of around 70°-100°C below 300°C, which CRIEPI used as the maximum acceptable temperature, this conclusion is open to argument. Extrapolating from this result and assuming a linear average seal heating rate, an 800°C fire would cause the seal to fail after approximately 2.5 hours. Exposure to higher temperatures would reduce the time to seal failure. Seal failure could also be induced by fires of lower temperature and longer duration. Furthermore, the synergistic effect of gamma radiation damage may lower the temperature threshold or reduce the time to failure at elevated temperature, a point which the Japanese did not consider when assessing the safety margin.¹⁵ Without an understanding of the probabilities with which different fire scenarios may be encountered during marine transport, it is impossible to judge whether a particular "safety margin" is sufficiently conservative.

b) Sensitization of stainless steel VHLW canisters

The stainless steel canister that encases VHLW plays an important role in ensuring the safety of transport, handling and storage of the material. It should be obvious that the chosen canister material should provide a high degree of confidence in its integrity. However, at the R7-T7 vitrification plant at La Hague and at the Waste Vitrification Plant (WVP) at Sellafield, the VHLW canister material being used does not provide such confidence. In fact, one can show that this material, known as Type SUH 309 austenitic stainless steel, undergoes a phase transformation while the VHLW canisters are being cooled. This phenomenon, known as *sensitization*, greatly reduces its resistance both to certain types of corrosion and to mechanical impact.

Sensitization of austenitic stainless steels occurs when the steel is held for a certain period of time at a temperature in the range of approximately 400-850°C. The time necessary to cause extensive sensitization depends on the composition of the steel, and in particular will decrease as the carbon content increases. For instance, one experiment found that while a 3 millimeter (mm) sample of stainless steel with a carbon content of 0.03% underwent 100% sensitization after 10 hours, one with a carbon content of 0.08% sensitized completely after only 30 minutes.¹⁶ Type SUH 309 has an even higher carbon content (0.15 weight-percent) and therefore will be completely sensitized in less time.¹⁷ Furthermore, it has been observed that stressed materials (such as the VHLW canisters) undergo sensitization more rapidly than unstressed ones.

COGEMA data shows that as the VHLW canisters are being cooled after being filled with glass, the canister temperature remains within the sensitization range for about 7 hours. Thus there is little doubt that the canisters being produced at La Hague are extensively, if not completely, sensitized.

A number of stainless steels have been developed which are resistant to sensitization. Partly for this reason, one of these steels, Type 304L, is being used as the canister material at the HLW vitrification plant now operating in the United States. Type 304L is also the steel that was chosen for use at the domestic HLW vitrification plants in Japan. However, one should note that even if Type 304L is used, sensitization may occur to a limited extent during VHLW production. (There are materials which are somewhat more resistant to sensitization, such as the high-nickel Alloy 825). *Why COGEMA and BNFL initially chose (and continues to use) a type of steel highly susceptible to rapid sensitization, apparently without prior consultation with Japanese authorities, is by no means clear.*

Austenitic stainless steels exhibit a high degree of resistance to *uniform* corrosion (corrosion that takes place uniformly across a surface). However, when they are exposed to certain chemical and thermal environments, they can undergo *localized* corrosion processes such as intergranular corrosion (IC) and intergranular stress-corrosion cracking (IGSCC).¹⁸ Localized corrosion can typically be two or three orders of magnitude more severe than uniform corrosion, and has often led to unexpected, catastrophic failures of materials.

Some contaminants, such as chloride salts and hydroxyl (OH⁻ ions), can initiate intergranular corrosion of stainless steels at very low concentrations, if water is also present. This is clearly a concern with regard to the integrity of VHLW canisters: for instance, when reporters were recently shown empty stainless steel canisters at the U.S. vitrification plant, they were warned not to touch them to prevent exposure of the steel to potentially corrosive salts in their sweat.¹⁹

Because the VHLW canisters being returned to Japan are produced, stored and shipped in marine environments (all facilities are located near oceans), the ambient air concentrations of chlorides from sea salt are always high and extreme care should be taken to prevent excessive salt contamination of the canisters. It is not clear that such care is taken, however.

Stainless steel that has become sensitized is much more vulnerable to localized, intergranular corrosion than the same type of steel in the unsensitized condition. Because localized corrosion is a

much more unpredictable phenomenon than uniform corrosion (e.g. it is more susceptible to small changes in environmental conditions) it is less accurate to apply the results of laboratory tests to predictions of long-term performance, or to extrapolate data from a single sample to an entire lot. Thus one consequence of using sensitized stainless steels in VHLW canisters is that uncertainties in predictions of canister performance will be greatly increased. This can only reduce confidence in the results of safety analyses.

In the event of an accident in which a VHLW cask is damaged and then lost at sea, as described above, the accelerated corrosion of sensitized Type 309 stainless steel in seawater will virtually guarantee that the canisters will fail within a few months' time, exposing the radioactive glass matrix inside to seawater.

Conclusions

The shipment of VHLW by sea is a practice with potentially catastrophic consequences for the inhabitants and economies of coastal states along the shipping route. In particular, the loss of a VHLW cargo ship in coastal waters, a credible event, can result in extensive radioactive contamination of the environment. However, the industry responsible for the shipments continues to refuse to provide to the public the technical basis for its assurances that it has reduced the probability of accidents to an acceptable level. In many cases, the technical basis does not exist. Given this situation, en-route states would be justified in not permitting the passage of these cargos through their Exclusive Economic Zones.

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End Notes

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2. See, for example, "Vitrified Waste on the High Seas," *SpentFuel*, January 30, 1995, p.3; "Shipment of Nuclear Materials from France to Japan," British Nuclear Fuels plc. media brief (undated). [Back to document](#)
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10. Central Research Institute of [the] Electric Power Industry, Japan, "Environmental Impact

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11. The cumulative (decay-corrected) exposure over the 50-year period from the three radionuclides is 0.73 Sv, of which 93% results from ingestion of alpha-emitting radionuclides. The appropriate risk factor to apply is therefore $(0.1 \text{ fatal cancer/Sv}) (0.93 + 0.5 \cdot 0.07) = 0.097 \text{ fatal cancers/Sv}$. This is because the risk associated with high linear energy transfer (LET) radionuclides, such as alpha-emitters, is about 0.1/Sv, while for low-LET radionuclides (such as Cs-137, a beta-gamma emitter), it is about 0.05/Sv. The fact that there is no evidence for applying a dose-rate effectiveness factor for high-LET radiation, as pointed out by the United States BEIR Committee, generally is ignored in risk calculations [National Research Council, U.S. National Academy of Sciences, *Health Effects of Exposure to Low Levels of Ionizing Radiation (BEIR V)*, National Academy Press, Washington, D.C., 1990, p.6). [Back to document](#)

12. The industry has denied that elastomer seals are a bad choice. However, they do not refute the argument that metallic seals would be far more likely to survive a severe fire. [Back to document](#)

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15. M. Warrant and C. Ottinger, "Compilation of Current Literature on Seals, Closures and Leakage for Radioactive Material Packagings," SAND88-1015, Sandia National Laboratories, January 1989, p.4-10. [Back to document](#)

16. H. Solomon, "Influence of Composition on Continuous Cooling Sensitization of Type 304 Stainless Steel," *Corrosion* 40 (1984) 51. [Back to document](#)

17. COGEMA now says that the carbon content of the Type 309 stainless steel that it is using has been lowered to 0.08%, rather than the maximum possible value of 0.15%. It is not clear whether this has always been the case or whether the alloy was changed recently in response to the work of this author. However, this point does not change any of the conclusions of this paper, because stainless steel with 0.08% carbon is also highly vulnerable to sensitization, as the above discussion makes clear. [Back to document](#)

18. Stress-corrosion cracking is a special type of corrosion that can only occur on a surface which is under tensile stress. Although there is no generally agreed-upon minimum stress threshold below which it cannot occur, it becomes more likely for stresses greater than about 70% of the yield strength of the material. As pointed out above, the stainless steel VHLW canisters experience tensile stresses in storage at or above their yield strengths. [Back to document](#)

19. M. Wald, "Factory is Set to Process Dangerous Nuclear Waste," *New York Times*, March 13, 1996, p. A16. [Back to document](#)

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