Station 5 JUL 14 1999	12	ENGINEERING DATA TRANSMITTAL	Page 1 of 1 1. EDT 624178
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2. To: (Receiving Organization) PFP Project		3. From: (Originating Organization) Plutonium Process Support Labs		4. Related EDT No.: NA	
5. Proj./Prog./Dept./Div.: PFP Project		6. Design Authority/Design Agent/Cog. Engr.: T. D. Cooper		7. Purchase Order No.: NA	
8. Originator Remarks: None				9. Equip./Component No.: NA	
				10. System/Bldg./Facility: 234-5Z	
				12. Major Assem. Dwg. No.: NA	
				13. Permit/Permit Application No.: NA	
11. Receiver Remarks:				14. Required Response Date: 07/13/99	
11A. Design Baseline Document? ☐ Yes ☒ No					

15. DATA TRANSMITTED					(F)	(G)	(H)	(I)
(A) Item No.	(B) Document/Drawing No.	(C) Sheet No.	(D) Rev. No.	(E) Title or Description of Data Transmitted	Approval Designator	Reason for Transmittal	Originator Disposition	Receiver Disposition
1	HNF-4829	NA	0	Technical Documentation	SQ	1	1	1
				to Support the Evaluation				
				of Handling of Plutonium				
				Metal				

16. KEY		
Approval Designator (F)	Reason for Transmittal (G)	Disposition (H) & (I)
E, S, Q, D OR N/A (See WHC-CM-3-5, Sec. 12.7)	1. Approval 2. Release 3. Information 4. Review 5. Post-Review 6. Dist. (Receipt Acknow. Required)	1. Approved 2. Approved w/comment 3. Disapproved w/comment 4. Reviewed no/comment 5. Reviewed w/comment 6. Receipt acknowledged

17. SIGNATURE/DISTRIBUTION (See Approval Designator for required signatures)											
(G) Reason	(H) Disp.	(J) Name	(K) Signature	(L) Date	(M) MSIN	(G) Reason	(H) Disp.	(J) Name	(K) Signature	(L) Date	(M) MSIN
		Design Authority									
1		Design Agent TD Cooper	<i>T.D. Cooper</i>	7/13/99	T5-12						
1		Cog. Eng. TD Cooper	<i>T.D. Cooper</i>	7/13/99	T5-12						
1		Cog. Mgr. CS Sutter	<i>C.S. Sutter</i>	7/13/99	T5-12						
1		QA DR Groth	<i>DR Groth</i>	7/13/99	T4-15						
1		Safety SE Nunn	<i>SE Nunn</i>	7/13/99	T5-11						
		Env.									

18. <i>T.D. Cooper</i> T. D. Cooper Signature of EDT Originator	7/13/99 Date	19. <i>D.R. Speer</i> D. R. Speer Authorized Representative for Receiving Organization	7/14/99 Date	20. <i>C.S. Sutter</i> C. S. Sutter Design Authority/Cognizant Manager	7/13/99 Date	21. DOE APPROVAL (if required) Ctrl No. _____ ☐ Approved ☐ Approved w/comments ☐ Disapproved w/comments
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TECHNICAL DOCUMENTATION TO SUPPORT THE EVALUATION OF HANDLING OF PLUTONIUM METAL

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P. O. Box 1200
Richland, WA 99352
U.S. Department of Energy Contract DE-AC06-96RL13200

EDT/ECN: 624178 UC:
Org Code: 15F00 Charge Code: 100741
B&R Code: Total Pages: 40

Key Words:

plutonium metal, plutonium hydride, plutonium storage

Abstract:

in 1997, a can containing a plutonium metal ingot was opened. The sides of the inner storage can had collapsed. As the inner can was opened, an apparent flame appeared to issue from the opening. Based on the reaction and possible pressurization of the glovebox, a positive Unreviewed Safety Question (USQ) screening was issued. This document contains some of the technical documents to resolve the screening.

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HNF-4829

TECHNICAL DOCUMENTATION TO SUPPORT THE
EVALUATION OF HANDLING OF PLUTONIUM
METAL

JULY 12, 1999

Revision 0

Introduction

In 1996, a can from the Plutonium Finishing Plant (PFP) vaults containing a plutonium ingot was opened. The plutonium within the can had consumed enough of the air in the inner storage can to cause the sides to collapse in a so-called "paneled" condition. As the electric can opener began opening the can, an apparent flame about the size of a person's hand appeared to issue from the opening that lasted for approximately 80 milliseconds. As the lid was removed, the remaining corrosion products and some spots upon the plutonium ingot glowed red for several minutes.

This display of pyrotechnic energy was higher than observed previously, and formed the basis for a concern that we might not have a complete understanding or appreciation for the reactivity of the stored metal. A 150 gram bounding estimate for plutonium hydride was formulated by transferring all of the hydrogen in one contamination control bag to the plutonium.

A literature review found the cloud combustion of 150 grams of plutonium hydride with air could result in an unacceptable pressurization of the glovebox. On this basis an Unreviewed Safety Question (USQ) screening was issued in 1997 concerning opening cans containing stored plutonium metal items.

Discussion

In 1999, T. D. Cooper issued three technical letters containing supporting information for resolving these safety issues. It was decided that incorporating these letters within an SD document was advantageous for insuring their retention, their retrievability, and ease of reference. These letters are therefore included as Appendices A, B, and C. The results from these attachments are summarized below:

Appendix A

Letter, T D. Cooper, BWHC, to R. W. Szempruch, BWHC, "Obtaining Closure on Reactive Metal", 15F00-99-044, dated May 7, 1999.

The Appendix A letter answers four safety concerns. The general conclusions in the letter are:

- 1) Hanford assumes leaky food-pack can seals and forms a plutonium hydride limit with the total amount of hydrogen within the contamination control bag. Savannah River Plant personnel assumed no leaking seals and bound their hydride limit as a result of radiolysis of the bag or reaction of the plastic with the innermost can atmosphere.
- 2) Hanford finds the reaction rate of the corrosion products bounded by 200 grams of plutonium hydride to be even though a very low probability may be

cited for the formation of larger amounts of plutonium nitride through a very slowly leaking container seal.

3) The Hanford contingent based their safety case on the historical record of handling 100-200 grams of plutonium hydride within an air-filled glovebox at Rocky Flats Environmental Technology Site (RFETS). It was questioned whether this case bounds the reactivity and total energy contained within potentially larger amounts of plutonium sesquioxide, nitride and/or metal fines. It was shown that plutonium hydride yields the highest energy dispersal rate and this rate forms the bounding safety case rather than the total reaction enthalpy.

4) A concern was expressed that exposure to multiple containment bags could increase the amount of hydride beyond the 150 gram limit obtainable from a single bag. It was shown in response that the hydrogen transfer rate is not linear with time but rather decreases exponentially as unsaturation increases within the plastic. Therefore exposure to multiple bags is not expected to increase the amount of hydride beyond that expected by transfer of all of the hydrogen from a single bag.

Appendix B

Letter, T. D. Cooper, BWHC, to M. W. Gibson, BWHC, "Calculations Bounding GB-21A Pressure and Temperature", 15F00-99-045, dated May 7, 1999.

The information contained in the letter is summarized by stating that the energy generation rate necessary to raise glovebox HA-21A from -0.5 inches of water to 0 inches of water is 14,665 J/sec. It was shown that the heat transfer rate to the glovebox air from a typical powder boat filled with powder at 1561 °C is twenty seven times less than that required to pressurize the glovebox.

Appendix C

Letter, T. D. Cooper, BWHC, to E. P. Bonadie, BWHC, "Scientific Evaluation of Safety in Processing Pu Metal", 15F00-99-054, dated May 25, 1999.

The objective of the letter in appendix C was to show clearly those safety cases that can be resolved rigorously and those that are being qualified through practical experience. Oxygen transfer through air to the corrosion powder surface was the first process to be evaluated. The second was intra-bed diffusion, and the third was intra-particle diffusion. We only have the data to evaluate the first process rigorously. The results show that heat transfer from an inner can is only 20% of that required to pressurize the glovebox. This limit is based upon the diffusion rates and convection rates for introducing oxygen into the inner can.

If the corrosion product is spread out in an open powder boat, the supply of oxygen to the surface does not form an acceptable limit. It was shown however in Appendix B that the heat transfer rate from a 1561 °C powder surface to the glovebox air is twenty seven times less than that required to pressurize the glovebox.

If the corrosion product is lofted into a cloud, no diffusional or heat transfer rate barriers are available to prevent pressurizing the glovebox.

Since performing intra-bed and intra-particle diffusion calculations that could further reduce the reaction rate requires research work to establish the desired diffusion parameters, and since such research is costly in terms of both time and money, it was decided to rely upon a hydride handling limit that could be defended historically. This limit was 200 grams of plutonium hydride from the RFETS experience.

Appendix A

Letter, T D. Cooper, BWHC, to R. W. Szempruch, BWHC, "Obtaining Closure on Reactive Metal", 15F00-99-044, dated May 7, 1999

B&W Hanford Co.

a McDermott Company

To	R. W. Szempruch	T5-48	
From	T. D. Cooper Plutonium Process Support Laboratories	T5-12	File No. 15F00-99-044 Or Ref.:
Subj	OBTAINING CLOSURE ON REACTIVE METAL PROCESSING		Date: May 7, 1999

- References:
1. Letter, R. W. Szempruch, BWHC, to C. S. Sutter, BWHC, "Request for Closure Documentation on Denver Workshop Consensus-Plutonium Hydride Issues", 15530-99-RWS-031, dated April 20, 1999.
 2. HNF-2061, Rev. 0, *"Gas Generation from Plastics During Vault Storage by Way of Thermolytic and Oxidation Reactions"*, T. D. Cooper, dated May 6, 1999.
 3. Letter, T. D. Cooper, BWHC, to M. W. Gibson, BWHC, "Calculations Bounding Glovebox 21-A Pressure and Temperature", 15F00-99-045, dated May 7, 1999.
 4. HNF-2062, Rev. 0, *"Gas Generation from Plastics During Vault Storage by Way of Radiolysis Reactions"*, T. D. Cooper, dated May 6, 1999.
 5. LA-UR-97-2592, Rev. 0, *"Oxidation Kinetics of Plutonium in Air: Consequences for Environmental Dispersal"*, J. M. Haschke, T. H. Allen and J. C. Martz.
 6. EGG-FSP-9435, Rev. 0, *"Preliminary Analysis of Uranium Bed Air-Exposure Test Data"*, G. R. Longhurst, dated December 1990.

The Denver Plutonium Metal Corrosion Workshop was held November 10 and 11, 1998 to provide a forum for expert scientists to discuss issues concerning handling reactive plutonium corrosion products. Based on this workshop, participants concluded that the Unreviewed Safety Question (USQ) evaluation of opening cans of metal should be replaced with one that finds that no USQ exists. Reference 1 raised four issues concerning the Denver Workshop results and requested further clarification.

Introduction

Before entering into specific discussions of each of the four issues raised in the referenced letter, it is desirable to ensure the reading audience has a firm grasp of the chemical principles controlling reaction rates.

Reaction rates are typically described by equations utilizing reagent concentrations and temperature. Special circumstances may move reaction rate control from concentrations and temperature to other factors. Diffusion controlled reactions are an example of special circumstance. In diffusion controlled reactions, one or more reagents are severely depleted such that the overall rate is no longer dependent on the inherent reactivity of a particular reagent but instead is controlled by the delivery rate of a second reagent to the reaction site. Stated slightly differently, a diffusion controlled reaction is not dependent upon the concentration of reagent A or its inherent reactivity. It is simply determined by the rate of diffusion of reagent B and its temperature dependence depends only upon the effect of temperature upon the diffusion rate of reagent B.

When "sparky" initiated this safety evaluation, it was obvious that we were dealing with very fast, energetic reactions. Because of a paucity of reaction rate information, our first approach was to try to limit the total reaction enthalpy to an amount that could be tolerated by the glovebox. If this approach was successful, we could say that the reaction rate was unimportant since the total energy was insufficient to challenge the glovebox. Calculations using total reaction enthalpy for plutonium hydride showed that if all of the reaction enthalpy were released instantaneously, 20 grams of plutonium hydride could raise the glovebox pressure from negative to positive. If all of the hydrogen from one containment bag were transferred to the plutonium metal, 150 grams of hydride could form and this amount could raise the glovebox internal pressure to approximately 3 psig. This pressure was high enough to challenge glovebox containment.

It was also shown experimentally with uranium hydride that if 150 grams were allowed to react in a dust cloud, the pressure wave reach 3 psig in an air volume similar to Glovebox HA-21A; thus experimentally validating the instantaneous pressure calculation. It should be noted that in a dust cloud, each particle is bathed in a reactive atmosphere and no bed-type diffusion control is possible.

Since insufficient knowledge existed at the beginning of this program to prove that all of the hydrogen from a single bag could not reach the Pu metal, 150 grams of Pu hydride or its equivalent was accepted as a bounding limit. Bounding in this sense does not mean that 150 grams of hydride is acceptable for our safety case, but simply that we did not expect more than 150 grams of hydride to form.

Reactivity modeling of the system using published hydride and nitride reaction rates was successful in demonstrating pyrophoric ignition of these systems. Such modeling, however, failed to bound the equilibrium reactivity since the system temperatures rapidly increased beyond those from which the measured reaction rates were derived and since the modeling was not constructed to account for bed-diffusion limitations.

Just before the Denver Workshop, David Horrel stated in a telephone conversation that 100 grams of Pu hydride had burned in air gloveboxes almost routinely at the Rocky Flats Environmental Technology Site (RFETS) and as much as 200 grams had been burned on occasion. This was proof that the reaction rate was diffusion controlled since otherwise glovebox pressurization would surely have occurred.

Because of the high surface area and high inherent reactivity, it is obvious that plutonium hydrides, nitrides, and metal fines can all be self igniting, and the reaction rate quickly becomes diffusion controlled in a powder bed. The reaction rate will then be diffusion controlled by the reactant gas concentration, the geometrical area of the powder bed, the size of the channels between particles, and the tortuosity of the channels within the bed. If hydride, nitride and metal fines all had a similar size distribution, one would predict that the bed characteristics would be uniform and, therefore, under diffusion control, the equilibrium reaction rates would be uniform.

We do not have a characterization of the corrosion product identities or particle sizes but one can make the argument that millimeter sized particles or larger should possess low specific surface areas that their reactivity will not challenge glovebox containment. To potentially react at a rate fast enough to challenge glovebox containment, the particles must be micron sized. The practical consequence argument therefore implies that such beds must burn with fairly uniform diffusion controlled rates and the chemical identity is not very significant.

One further predicts that when the bed depth becomes sufficient to establish diffusion control, increasing the mass within the bed does not increase the reaction rate. It simply means that the bed will burn for a longer time. One further predicts that once diffusion control is achieved and if all other factors are equal, a metal bed will produce the maximum heat generation rate, a hydride bed will produce the second highest heat generation rate, and a nitride bed will produce the lowest. This order is determined by the reaction enthalpy of oxygen with each of the substrates.

With this understanding, it is obvious that our safety case must be built upon the fact that reaction of the metal corrosion products must be kept in a diffusion controlled regime and that dust clouds must be avoided. The best diffusion control is achieved in a bed of particles.

With this basic foundation, we will now discuss the four issues raised in Reference 1.

Discussion of Four Issues Raised

1. *Of particular concern from the Denver Workshop is the presentation by Major Thompson relative to its applicability to most of the cans of metal currently stored at PFP. If I understand the presentation, 10 grams of polyethylene generates 359 ml hydrogen in 20.8 years. Five grams of polyvinylchloride generates 840 mL of hydrogen in the same time period. This data indicates that for the nominal 20-30 gram PVC bags plus tape used to package our metal over 30 years ago that a more appropriate estimated of the hydrogen generated by decomposition of the bag would be at least 4200 mL. This is about four times the quantity used by Major Thompson to show the quantity of plutonium hydride that might be formed in a sealed can. Thurman Cooper's recent draft document (HNF-2062 Rev. 0, Gas Generation from Plastics During Vault*

May 7, 1999

Storage by Way of Thermolytic and Oxidation Reactions) reinforces my concern in that it reports that polyvinylchloride is 98 percent dehydrochlorinated within 30 years.

Response:

Major Thompson calculated the hydrogen release from polyethylene as it oxidized to carbon monoxide, carbon dioxide (CO_2), and water (H_2O). Major limited the amount of oxygen to that contained originally in the container thus ignoring any oxygen leaking into the container through the seal. Major's hydrogen release estimate is therefore dependent upon the amount of oxygen available to the plastic and independent of the amount of plastic in the container. If one applies Major's hydrogen production rate to larger amounts of plastic assuming a constant ratio of oxygen to plastic, one can indeed calculate larger amounts of hydrogen released as water.

At the Denver Workshop, Major gave an estimate of 60 grams of plutonium hydride. This estimate was based upon both radiolytic and thermal-oxic degradation of the plastic in a sealed container. At Hanford, our initial estimates for the amount of plutonium hydride to be found in a plutonium metal ingot storage container ranged from 0 to 40 grams based upon prior handling experience. Since no analytical data for hydride in the corrosion product exists, the Hanford contingent adopted the entire 0.622 g-moles of hydrogen in the plastic contamination control bags as a basis for calculating a plutonium hydride limit. This correlates to 150 grams of PuH_2 .

Just before the Denver Workshop, an attempt to limit the bounding hydride estimate to a lesser value was launched at Hanford. It was intended to model the radiolytic and thermal release of hydrogen from the contamination control bag. At the time of the Denver Workshop, radiolytic modeling at Hanford yielded enough hydrogen to form 60 grams of plutonium hydride. Since Savannah River Technology Center (SRTC) and Hanford both had dissimilar models yielding 60 grams of hydride, this amount was discussed at the Denver Workshop as a potential hydride limit but it was quickly recognized that the available evidence was not strong enough.

The Denver Workshop recommendations were intended to cover cases where the total amount of hydrogen in the plastic transfers to the metal and up to the maximum amounts of hydride that were handled in air filled gloveboxes at RFETS and at Los Alamos National Laboratory (LANL).

Since the Denver Workshop, Thurman Cooper continued to perform radiolytic and thermolytic modeling of polyethylene and PVC. The hydrogen yield from radiolytic modeling was decreased by more than a factor of two once a decreasing G value for hydrogen production was incorporated into the model. These modeling results are given in Reference 2.

For thermolytic-oxic plastic degradation, it is realized that the release of hydrogen most likely follows an exponentially decreasing curve with time. As plastic is oxidized, its chemical characteristics change and it becomes increasingly difficult to extract hydrogen from the plastic.

Data for the thermolytic and oxidation model was obtained from short-term experiments. The longest experiment recorded was 15 days. Because of lack of long-term hydrogen release data, the short-term release rates were used to model long-term hydrogen releases. This admittedly conservative method allows a release estimate of 98% of the 0.622 g-moles of H_2 contained in a plastic bag during a 30 year storage interval.

There is no reason to believe that all of this hydrogen reaches the Pu metal since it can leak or diffuse outward as well as inward however a bounding assumption is that it all leaks inward thereby reacting with the metal to form 150 grams of plutonium hydride. This concern is unique to Hanford since the majority of our metal is contained in PVC, whereas SRTC Pu metal is contained in polyethylene.

In summary, the Hanford contingent does not accept Major Thompson's primary modeling assumption that the amount of hydrogen emitted from the plastic is limited to the amount of oxygen initially in the container. We believe that leaks in food pack can seals can increase the amount of oxidation to that required to potentially release all of the hydrogen in the plastic. We further believe that the crucial safety basis question is not the amount of reactive corrosion product that is formed but whether it reacts under powder bed diffusion control.

2. *As I read Jerry Stakebake's contribution provided after the workshop, addressing Consensus Item #9, I find that the RFETS experience supports my own observations that more than 60 grams of reactive corrosion products have been formed in stored metal items. Stakebake's conclusion is that the modeling discussed in the workshop does not bound what has been observed. This dichotomy with Consensus Item #9, "Sealed cans bound leaky cans for reactive corrosion products in air atmosphere" from the workshop needs to be reconciled.*

Response:

The Denver Workshop participants were focused upon showing that the maximum amount of plutonium hydride that could be created from a single bag was in the same range as the amount of hydride that had been burned at RFETS in an air glovebox. Jerry Stakebake's concern is that a slow air leak could passivate hydride and also create nitride thereby potentially creating a more severe bounding case than 150 grams of pure plutonium hydride.

If one assumes the initial formation of 150 grams of hydride, one of our typical metal ingots, approximately 20 grams will be attached to the ingot metal surface. The rest will have necessarily spalled. If one postulates that development of a slow leak, the spalled hydride reacts with oxygen and nitrogen, simultaneously releasing water and hydrogen gas. Since the metal is saturated with hydrogen and a hydrogen diffusion gradient exists, gas will tend to diffuse out of the innermost can and be lost to the system. The hydride in and on the metal will continue to catalyze the reaction of the metal.

The innermost can has a height of 7.30 cm and a radius of 3.33 cm with calculated volume of 255 cubic centimeters. A typical ingot volume is 146.6 cubic centimeters, giving a net headspace of 108.3 cubic centimeters. Since the volume of the ingots may vary, the associated headspace may also vary. The bounding headspace volume is therefore considered to be the innermost can volume.

If one assumes a nominal bulk density of 3 grams/cc for the corrosion product and a ratio of sesquioxide to nitride of 1/5.2 as reported by Haschke and co-workers, one may calculate the following:

Table 1 - Calculation of Characteristics of Corrosion Product

Cc Head Space	Bulk Density	Grams Product	g Oxide/ g Total	g Oxide	g-m Oxide	g Nitride	g-m Nitride
256	3	768	0.4	307	0.58	461	1.82

Reaction of this amount of plutonium nitride and sesquioxide with oxygen upon subsequent opening of the container yields 1378 and 91 kJoules respectively. In comparison, the reaction of 150 grams of hydride yields 748 kJ of energy.

The next question is just how rapidly this nitride-sesquioxide mixture reacts and how effectively it transfers heat energy to the glovebox. As discussed in the introduction, the reaction rates of hydrides and nitrides in a bed are limited by intra-particle and intra-bed oxygen diffusion rates. If the gaseous reaction rate becomes diffusionally limited and, for beds with similar cross-sectional area and powder characteristics, the same oxygen reaction rate will be observed in either a hydride bed or a nitride bed. In this case, the nitride bed will generate a smaller energy flow to the glovebox than the hydride bed due to the smaller oxygen-nitride reaction enthalpy.

The glovebox air pressure and temperature are functions of the initial conditions and the energy flow rate. Nitride is seen therefore to be less of a bounding hazard than hydride. It is recognized that if the nitride bed is more massive than the hydride bed, it will burn longer and may yield more total energy.

Plutonium corrosion products on metal have been observed to burn in air gloveboxes on many occasions. The red to yellow color on the burning surface indicates surface temperatures typically in the 800-1200°C range. Pure plutonium hydride in thermally isolated containers has occasionally reached surface temperatures as high as the melting point of platinum (i.e. as high as 2000°C). There is no evidence that any of our systems contain pure plutonium hydride or that the containers are thermally isolated. We therefore believe reasonable bounding surface temperature for our corrosion products is 1561K. Based upon this bounding surface temperature, a heat transfer model as described in Reference 3 shows that neither nitride nor hydride in a typical reaction boat will pressurize or overheat glovebox HA-21A.

This theoretical conclusion is further buttressed by the practical handling experience at RFETS.

3. *The total energy that may be released by reaction of all corrosion products (hydride, sesquioxide, nitride and metal fines) needs to be addressed. While calculations are certainly bounded if the hydride oxidation heat of reaction is substituted for the other oxidation reactions on a gram for gram basis, it is not sufficient to examine only the energy released from oxidation of the hydride while disregarding other reactions. Of most concern is the potential for metal fines that are formed during metal corrosion.*

Response:

The plutonium metal reaction with water vapor in the air produces a mixture of oxides and hydrides. The hydrides react further with oxygen to produce oxides and hydrogen is recycled catalytically to the metal as fresh hydride. The oxide scale adheres tightly until it thickens to a few nanometers and then begins to separate from the metal surface due to strains between the oxide crystal structure and the metal crystal structures. The oxide therefore has a leaf structure that continues to build in thickness until it reaches approximately 100 microns, whereupon it begins to spall from the metal. The spalled corrosion product resulting from air reaction contains unreacted hydride in the range of 1 to 5 weight percent depending upon the oxygen to water ratio and the temperature. The residual hydride from reaction with pure water vapor in a sealed system has been measured as high as 42 weight percent. High temperatures and high oxygen/water ratios favor complete reaction of the hydride, conversely low temperatures and low oxygen/water ratios favor high residual hydride concentrations.

Subsequent reaction of hydride in the spalled corrosion product with air at very low oxygen concentrations may allow the formation of nitrides and sesquioxides. These compounds contain less energy and possess slower reaction rates with oxygen than hydrides and, therefore, are bound in a safety sense by assuming all of the hydride remains as hydride. Subsequent catalytic reaction of the hydride dissolved at the metal surface may create large amounts of nitride and/or oxide.

The spalled corrosion product also contains unreacted micron-sized metal fines in the range of 1 to 5 weight percent. Since only a small portion of the corrosion products is expected to be micron-sized metal, this is believed to be a second order concern that does not significantly alter the bounding limit for reactivity.

Intergranular corrosion may cause chunks of metal with largest dimensions ranging from a millimeter to several centimeters to break free from the parent ingot. These large chunks of metal may contain considerable mass and, thus, increase the metal/oxide ratio but their specific surface area is so low as to not allow for appreciable reactivity. One may therefore argue that metal does not appreciably increase the reactive surface area and, secondly, that the entire system is diffusionally controlled rather than mass controlled. The presence of metal is not therefore expected to significantly perturb the energy transfer rate to the glovebox air.

4. *Within the UK metal category there are 10 different container configurations involving two, three and four can arrangements and nearly again as many for our other metal items. Most of these cans had the two outer sealed cans applied years after the inner can-bag-slip lid can configuration came into existence. Some have been repackaged in new plastic bags without opening the inner can containing the metal ingot. It is not clear to me that the participants of the Denver workshop recognized these situations.*

Response:

Denver Workshop participants discussed the fact that many packaging configurations exist and that these different packaging configurations definitely affect the actual amount of corrosion occurring on the metal. It was also discussed that one could not rule out the possibility of two bags being stuffed into a single can or that repackaging could have occurred on some of the cans. Since no one at the Denver Workshop could quantify the numbers or probabilities of multi-bag overpacks and since everyone agreed that the single bag case was dominant, the single bag case was the baseline for discussion at the workshop.

Due to the fact that hydrogen release is not linear over time as was assumed in Reference 4 and only half of the hydrogen is actually expected to be released from a single bag; one can make the argument that exposure to two bags sequentially is not expected to increase the bounding hydride case beyond the original 150 gram estimate.

If one couples the above argument with the fact that energy release is diffusionally controlled, one can reconcile the experimental observations that no one in the Department of Energy (DOE) complex has ever opened a Pu metal containing food pack can that exhibited enough corrosion and reactivity to challenge the glovebox containment. This statement is true for metal that has been exposed to both single bag and multi-bag configurations. It is therefore still our view that the hydride obtainable from a single bag is a reasonable bounding limit for multi-bag configurations and, that furthermore, if this bound were somehow exceeded by a low probability event, there is no evidence that glovebox containment is jeopardized.

5. *Technical papers and calculations were identified as part of the ten action items in the workshop notes. To the best of my knowledge most have not been completed. Without their completion the efforts of the Denver Workshop remain incomplete. In addition to workshop notes that may be referenced, these technical reports need to be available to the personnel performing safety evaluations of opening metal items.*

Response:

A review of the commitments made at the Denver Workshop for incomplete action items is given below:

8. Plutonium corrosion products collection will have a specific limit. (Corrosion products from one storage container or the accumulation is limited to that specified by safety assessment.)

Action: Publish Major Thompson's and Thurman Cooper's calculation documents

Thurman Cooper has issued his two documents, References 2 and 4. Major Thompson's document has been received and has been made available to PFP Engineering.

9. Sealed cans bound leaky cans for reactive corrosion products in an air atmosphere.

Action: Mark Bronson, Jeff Schaade, and Jerry Stakebake collaborate to produce white paper on the subject.

Jerry Stakebake published a white paper on this subject. His conclusions are contrary to item 7 and his conclusions are accepted by this author. It is further concluded that the reactivity of the corrosion products potentially obtainable from leaky cans is nevertheless bounded to acceptable consequences in the glovebox because of oxygen diffusion and heat transfer phenomena. (See the introduction remarks and the response above for Item 2.)

10. No credible mechanism exists for the reactive species in the can to cause glovebox over-pressurization by cloud dispersion.

Action: Publish Thurman Coopers's and Major Thompson's calculation documents.

Thurman Cooper's calculations address the thermal energy released by the oxidation of hydride. They do not address any mechanism for imparting kinetic energy to the particles. The rejecting of the formation of a hydride cloud has to be the simple fact that no mechanism has been identified for violently expelling fresh hydride into the glovebox and the lack of any historical basis for the formation of such a cloud.

The fact that no cloud formation is expected is best established through process history.

The Denver Workshop documents this process history through the collective experiences of the participants and their collective agreement that no such cloud has been seen experienced, or postulated.

The intent of the item has been met.

Action: Thurman Cooper to document the experiences.

Thurman Cooper has contacted David Horrell and discussed his experiences with hydride in air-filled gloveboxes and the lack of explosive cloud formations. This discussion supported information already provided.

11. A source of hydrogen and water to form hydride (PuH_2) is generated from both thermal and radiolytic degradation of plastic.

Action: Publish Thurman Cooper's and Major Thompson's calculation documents.

Thurman Cooper's two documents, References 2 and 4, have been issued. Major Thompson's document has been received and has been made available to PFP Engineering.

12. The thermal degradation of plastic material in plutonium storage containers dominates over radiolytic degradation when plastic is not in direct contact with plutonium materials.

Action: Richard Mason to talk with Gary Eller and Lav Tnadon and supply documentation.

Richard Mason has agreed to provide information on this subject. Due to Richard's other responsibilities, this information is not current available. Since Reference 2 and 4 reached the same conclusion, the work will serve as the basis until the contribution is received.

13. When PuH_2 oxidation occurs at a low temperature (not burning) the H_2 will be diluted by glovebox airflow.

Action: Thurman Cooper to provide derivation.

This action is satisfied since Haschke's work, Reference 5, and Longhurst's work, Reference 6, show that both PuH_2 and UH_3 do not emit hydrogen when exposed to air. Water and the metal oxide are the products.

14. When burning plutonium hydride, the hydrogen will oxidize as it evolved in an atmosphere that is not oxygen limited.

Action: Thurman Cooper to find supporting documentation.

This action is satisfied since Haschke's work, Reference 5, and Longhurst's work, Reference 6, show that both PuH_2 and UH_3 do not emit hydrogen when exposed to air. Water and the metal oxide are the products.

15. "Massive" plutonium metal button will not burn fast enough to cause safety problems. (RFETS uses ~50 grams as "massive". That is not to mean that less than 50 grams is a problem.)

Action: Publish Thurman Cooper's calculation on this subject.

Thurman Cooper performed some calculations last year and these were made available to engineering in draft form for the combustion of normal and hydrided Pu metal. These calculations do not have sufficient independent merit to deserve formal publication since their basis was Haschke's published reaction rates.

Haschke (Reference 5) prepared severely hydrided Pu ingots that completely burned in less than 20 minutes. From this burn, a $0.613 \text{ g Pu/cm}^2/\text{min}$ reaction rate was calculated. This reaction rate was an order of magnitude higher than observed for non-

hydrided metal, but did not pressurize the glovebox. His published reaction rates are available. Some have questioned whether faster reaction rates are possible for massive metal than Haschke's published rates.

PPSL chemists believe the Hanford Pu metal will not oxidize as fast as Haschke's metal for two reasons:

1. Haschke established ideal hydriding conditions for his ingots using temperature cycles to crack the oxide coating under vacuum and finally back filling with 1 atmosphere of pure hydrogen. As a consequence, his metal was severely hydrided both externally and internally.

The Hanford Pu metal has not been exposed to thermal cycling under vacuum. The hydrogen supply and pressure has been very limited. and the corrosion product typically contains a mix of oxide and hydride. There is no historical indication from handling any of the Hanford metal that it is internally hydrided or that it will burn at an accelerated rate even close to that observed by Haschke.

2. Secondly, to burn at a rate significantly faster than that recorded by Haschke, the intra-bed diffusion control limit would have to be exceeded. This intra-bed diffusion control can be changed or eliminated if the oxide particles are agitated or lofted into a dust cloud. There is no mechanism envisioned for agitating or lofting the oxide during the burning of Pu metal at Hanford.

For particles burning in a static pile, the oxide created by the burning process is not being removed and it continues to limit the access of oxygen to the underlying metal. Under these conditions, faster burning of metal at Hanford than that recorded by Haschke is not considered likely.

This discussion provides the closure on the issues identified in Reference 1. If you have any questions on the information provided, please contact T. D. Cooper on 373-2543.

Appendix B

Letter, T. D. Cooper, BWHC, to M. W. Gibson, BWHC, "Calculations Bounding GB-21A Pressure and Temperature", 15F00-99-045, dated May 7, 1999.

CORRESPONDENCE DISTRIBUTION COVERSHEET

Author
T. D. Cooper

Addressee
M. W. Gibson

Correspondence No.
15F00-99-045

Subject: CALCULATIONS BOUNDING GB-21A PRESSURE AND TEMPERATURE

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To	M. W. Gibson	T5-55	
From	T. D. Cooper <i>T.D. Cooper</i>	T5-12	File No. or Ref.: BWHC-15F00-99-045
Subj	CALCULATIONS BOUNDING GB-21A PRESSURE AND TEMPERATURE		Date: May 7, 1999

Introduction:

Metal Stabilization was halted at PFP after observing for the first time a flash of fire emanating from a metal storage can being opened. This particular storage can, unofficially labeled "Sparky", resulted in an Unresolved Safety Question (USQ). The original event did not yield unacceptable consequences, since containment was not compromised, and no one was injured. It was, however, an indicator that conditions within the storage can were more severe than previously observed and raised the question of whether we properly understood the bounding consequences.

Our safety envelope for glovebox operations requires that we do not lose containment. In addition to the requirement that no breach occurs, such as loss of gloves, seal bags, or windows, it is highly desirable to keep the pressure differential between the room and the glovebox negative, so that contaminants do not leak past gaskets and seals. In addition, the glovebox temperatures must not exceed operational limits and the workers must not be injured or personally contaminated.

To answer these safety questions, it is necessary to understand in depth the reaction characteristics of plutonium metal and its reactive corrosion products.

Description of Reactive Corrosion Products

Although no analytical evidence established the chemical identity of the reactive substances in "Sparky", the reaction products of plutonium with atmospheric and organic degradation gases are PuOH, Pu₂O₃, PuO₂, PuN, PuC, and PuH_{2+x}. Finely divided PuOH, Pu₂O₃, PuN, PuC, and PuH_{2+x} have all been shown to be pyrophoric, (self-igniting at room temperature). Since the radiolysis and thermolysis products of the plastic bag, used for contamination control, contain hydrogen, and since the presence of at least small amounts of hydride are required to catalyze the nitrogen reaction at the vault storage temperature, it is highly probable that hydrides are present in the reactive mixture. Since the reaction and energy production rates of all the reactive components are bounded by the reactivity of PuH_{2+x}, hydride will be used as a single component to bound the system reactivity.

Ideas for Bounding the Corrosion Product Mass

The first attempt to bound the reactivity of the system involved transferring all of the hydrogen within a given storage container to the plutonium metal thereby creating 150 grams of plutonium hydride. Even this amount of hydride yields unacceptable pressure excursions in glovebox 21-A if allowed to react instantaneously.

Further consideration shows that plutonium hydride can act as a catalyst for the reaction of both oxygen and nitrogen with plutonium metal. It is possible for plutonium to maintain a very low oxygen concentration within its storage environment, even in the presence of a slow atmospheric leak, thus allowing reaction of both oxygen and nitrogen. The resulting nitride-oxide mixture is only limited by the leak rate and the headspace within the storage cans.

If one assumes the initial formation of 150 grams of hydride, approximately 20 grams will be attached to the ingot metal surface. The rest will have necessarily spalled. If one postulates the development of a slow leak, the spalled hydride may react with oxygen and nitrogen releasing water and hydrogen gas. Since the metal is saturated with hydrogen, and a hydrogen diffusion gradient exists, gas will tend to diffuse out of the innermost can and be lost to the system. The hydride in and on the metal will continue to catalyze the reaction of the metal.

The innermost can has a height of 7.30 cm and a radius of 3.33 cm with a calculated volume of 255 cubic centimeters. A typical ingot volume is 146.6 cubic centimeters, giving a net headspace of 108.3 cubic centimeters. Since the volume of the ingots may vary, the associated headspace may also vary. The bounding headspace volume is therefore considered to be the innermost can volume.

If one assumes a nominal bulk density of 3 grams/cc for the corrosion product, and a ratio of sesquioxide to nitride of 1/5.2 as reported by Haschke and co-workers, one may calculate the following:

Table 1- Calculation of Characteristics of Corrosion Product

Cc Head space	Bulk Density	Grams Product	g Oxide/ g Total	g Oxide	g-m Oxide	g Nitride	g-m Nitride
256	3	768	0.4	307	0.58	461	1.82

Reaction of this amount of plutonium nitride and sesquioxide with oxygen upon subsequent opening of the container yields 1378 and 91 kJ respectively. In comparison the reaction of 150 grams of hydride yields 748 kJ of energy.

From the perspective of total energy yield we thus see that filling the innermost container headspace with nitride and sesquioxide, becomes the bounding case.

If the head space of the inner storage can is filled with plutonium nitride, the nitride-air potential reaction enthalpy becomes bounding. Since the published nitride-air reaction rates are several orders of magnitude lower than the hydride-air reaction rates, at ambient temperature, the practical consequences of exposing 150 grams of hydride to air are still believed to be the bounding safety case.

Diffusional Kinetics

Information was received from David Horrell at Los Alamos National Laboratory (LANL), that while he was at RFP, they routinely burned from 50 to 200 grams of plutonium hydride in an air glovebox without serious incidents or consequences. In addition, he stated that at no time did he observe a corrosion product dust cloud formation or explosion, and neither were any secondary gaseous combustion events observed. This is clear experimental evidence that dust clouds are not likely to form, the burning rate of a hydride bed is not instantaneous, and is limited by diffusional processes and combustible mixtures of hydrogen gas do not separate from the burning surface without immediate combustion.

To provide proper bounds for the overall combustion rates, heat evolution rates and surface temperatures, it thus became necessary to understand the reaction rates, gaseous diffusion rates, and heat transfer mechanisms within the system. The first clue to understanding the reaction rate lies in the demonstrated ability of hydride to burn at the diffusion limit. Burning at the diffusion limit means that the burning process is controlled by the rate at which oxidant is supplied to the solid surface rather than the temperature of the solid surface. It also means that reaction of the oxidant typically occurs with very high probability when the oxidant touches the solid surface.

Under these conditions, as soon as the hydride reaction rate becomes diffusionally controlled, the penetration of oxygen is limited to a layer near the surface. The temperature is determined by the balance between heat generation in the surface layer and heat dissipation within the bed and from the bed to any available heat sink. In a bed with a good heat sink, the hydride glows red at the surface indicating the temperature ranges from 700 to 1000 °C. If the bed is thermally isolated, so that conduction cannot occur, temperatures in excess of 2000 °C have been observed.

As the surface layer is oxidized, penetration of oxygen into the bed proceeds until combustion is occurring within the bed interior. Combustion deep within the bed is further slowed by intra-bed diffusional limitations. When hydride beds are exposed to air, combustion typically takes several minutes and a bed of oxide results. The presence of experimentally observed beds of loose friable oxide is evidence that the combustion temperature did not exceed the 2400 °C melting point of the oxide product.

Establishing a Glovebox Pressure Bound

The independent parameters necessary to calculate the energy flow required to pressurize Glovebox HA-21A are contained in Table 2.

Table 2- Independent Parameters for Pressurization Calculation

Gb Pressure Atm*	Gb Volume - Liters	Gb Flow Rate - LPS	Gb Temp - K
0.998771	2947	16.5177	298.15

* Minimum Operational Pressure

The energy required to pressurize the glovebox may be calculated from the following:

$$\begin{aligned} dn/dt &= 16.5177 \text{ LPS} / 24.464 \text{ L/g-m} \\ &= 0.675 \text{ g-m/sec} \end{aligned} \quad (1)$$

One must now find an equivalency between dn/dt and dQ/dt

$$\text{for given } V, T \quad dn/dt = (P \cdot V/R) \cdot d(1/T)/dt \quad (2)$$

The change in temperature with time may be calculated from Table 3

Table 3- Glovebox 21-A Heating Rate

Pressure - atm.	Volume Liters	Moles Gas	Gas Constant	$d(1/T)dt$
0.998771	2947	120.46272	0.08205	1.882E-05

So $dT/dt = 1.682$ degrees Celsius per second

From the first law of thermodynamics

$$\Delta E = Q_2 - W_2 \quad (3)$$

This may be written as an average rate equation over the time interval δt by dividing by δt

$$\Delta E/\delta t = \delta Q/\delta t - \delta W/\delta t \quad (4)$$

During the time interval δt , gases (δM_i) enter the glovebox, (δM_e) leave the glovebox,

The addition and subtraction of the flow mass represents a change in the internal energy of the system which can be represented by ($e_i \delta M_i$) and ($e_e \delta M_e$) respectively.

dQ enters the system through either the system boundary or the release of chemical energy.

As heat dQ enters the system, work is also performed by the expansion of the system gases. The initial system is defined as representing all of the gases within the glovebox at time 0 and all of the gases (δm_i) that will enter the glovebox within the time δt .

$$\text{with these definitions, } \Delta E = E_2 - E_1 \text{ wherein } E_2 = E_{t+\delta t} + e_e \delta M_e \text{ and } E_1 = E_t + e_i \delta M_i \quad (5)$$

$$\text{therefore } \Delta E = E_2 - E_1 = (E_{t+\delta t} + e_e \delta M_e) - (E_t + e_i \delta M_i) = (E_{t+\delta t} - E_t) + (e_e \delta M_e - e_i \delta M_i) \quad (6)$$

$$\text{Now } \delta M_e - \delta M_i = [\sum_A \rho V_r \cos \alpha dA]_{av} \delta t \quad (7)$$

Σ = summation sign

ρ = gas density

V_r = Gas velocity

$\cos \alpha$ = gas flow fraction = $(M_e - M_i)/M_e$

A = cross section through which gases are flowing

δt = time interval

$$\text{so } (e_e \delta M_e - e_i \delta M_i) = [\sum_A \rho V_r \cos \alpha dA]_{av} \delta t \text{ and } (e_e \delta M_e - e_i \delta M_i)/\delta t = [\sum_A \rho V_r \cos \alpha \delta A]_{av} \quad (8)$$

$(E_{t+\delta t} - E_t)$ represents the change in energy within the control volume during δt

$$E_{c.v.} = \sum_m e \delta m = \sum_v e \rho \delta v \quad (9)$$

wherein dv represents an element of volume in the control volume, and the integration is over the

entire control volume.

$$(E_{t+\delta t} - E_t) = \delta \sum_m e \delta m = \delta \sum_v e \rho \delta v \quad (10)$$

the average rate of energy change within the control volume is then

$$(E_{t+\delta t} - E_t)/\delta t = \delta/\delta t \sum_m e \delta m = \delta/\delta t \sum_v e \rho \delta v \quad (11)$$

$$\text{so } \Delta E/\delta t = \delta/\delta t \sum_v e \rho \delta v + [\sum_A \rho V_r \cos \alpha \delta A]_{av}$$

(12)

in analyzing the work done by the system, it is convenient to distinguish between the work done

on the masses δm_i and δm_e as they enter and leave the control volume, and $W_{c.v.}$

Work is done by the normal force acting on δm_i and δm_e , as these masses cross the surface

let the normal tensile stress = $-\sigma_n$ then:

The normal force is defined by the tensile stress multiplied by the area dA .

work is then defined as the force acting through a distance dl and:

$$\delta W = -\sigma_n * \delta A * \delta l \quad (13)$$

Letting the system pressure P = the normal tensile stress

$$\delta W = \delta W_{c.v.} + [P_{eve}\delta M_e - P_{ivi}\delta M_i] \quad (14)$$

this can be rewritten as a surface integral

$$W = \delta W_{c.v.} + [P_v \rho V_r \cos \alpha \delta A]_{av} \delta t \quad (15)$$

$$\text{and so } \delta W / \delta t = \delta W_{c.v.} / \delta t + [\sum_A P_v \rho V_r \cos \alpha \delta A]_{av} \quad (16)$$

Now combining elements of the above equations

$$\delta Q / \delta t = \delta / \delta t \sum_v e \rho \delta V + [\sum_A e \rho V_r \cos \alpha \delta A]_{av} + \delta W_{c.v.} / \delta t + [\sum_A P_v \rho V_r \cos \alpha \delta A]_{av} \quad (17)$$

combining the two surface integral terms

$$\delta Q / \delta t = \delta / \delta t \sum_v e \rho \delta V + [\sum_A (e + P_v) \rho V_r \cos \alpha \delta A]_{av} + \delta W_{c.v.} / \delta t \quad (18)$$

The elements of the above equation are defined as follows:

$\delta Q / \delta t$ = the heat flow rate into the system

$\delta / \delta t \sum_v e \rho \delta V$ = the rate of change of the internal energy of the control volume

$[\sum_A (e + P_v) \rho V_r \cos \alpha \delta A]_{av}$ = the energy equiv. of the flow mass difference and the flow mass work

$\delta W_{c.v.} / \delta t$ = the work performed by a change in the control volume

Now $\lim \delta Q / \delta t = dQ_{c.v.} / dt$ as $\delta t \rightarrow 0$

$$\lim \delta / \delta t \Sigma v_e p \delta V = d / dt \Sigma v_e p dV \text{ as } \delta t \rightarrow 0$$

$$\lim [\Sigma_A (e + Pv) \rho V_r \cos \alpha \delta A]_{av} = [\Sigma_A (e + Pv) \rho V_r \cos \alpha dA] \text{ as } \delta t \rightarrow 0$$

$$\lim \delta W_{c.v.} / \delta t = dW_{c.v.} / dt \text{ as } \delta t \rightarrow 0$$

$$dQ_{c.v.} / dt = d / dt \Sigma v_e p dV + \Sigma_A (e + Pv) \rho V_r \cos \alpha dA + dW_{c.v.} / dt \quad (19)$$

These rates can then be integrated as a function of time if the relationship is known.

Now to evaluate these integrals

$$de / dt = 1.682 \text{ oC/sec} * 20.61 \text{ J/mole-oC} = 34.666 \text{ J/mole-sec}$$

$$de / dt = 34.666 \text{ J/mole-sec} / 28.8 \text{ g/g-mole air} = 1.2037 \text{ J/g-sec}$$

$$\Sigma v_e p dV = 2947 \text{ Liters} * 1.191 \text{ g/L} = 3511.02 \text{ grams}$$

$$(d / dt \Sigma v_e p dV) = 1.2037 \text{ J/g-sec} * 3511.02 \text{ grams} = 4226.157 \text{ J/sec}$$

$$\text{Work of expansion} = \Sigma v_e P dv = 1 \text{ atm} * (0) = 0 \text{ liter-atm/sec} = 0 \text{ J/sec}$$

$$\text{J/g-mole (at 298.15 K)} = 20.601 \text{ J/g-mole/oC} * 298.15 \text{ oC} = 6142.188 \text{ J/g-mole}$$

$$\text{J/g (at 298.15 K)} = e = 6142.188 \text{ J/g-mole} / 28.8 \text{ g/g-mole} = 213.27 \text{ J/g}$$

$$\text{Now to evaluate } \Sigma_A (e + Pv) \rho V_r \cos \alpha dA$$

By rearranging equation 19

$$\Sigma_A (e + Pv) \rho V_r \cos \alpha dA = dQ_{c.v.} / dt - d / dt \Sigma v_e p dV - dW_{c.v.} / dt$$

Now if we choose an energy flow rate of 10000 J/s

$$\Sigma_A (e + Pv) \rho V_r \cos \alpha dA = 10000 \text{ J/s} - 4226.157 \text{ J/sec} - 0 = 5773.8433 \text{ J/sec}$$

When the glovebox heats enough to raise the glovebox pressure to atmospheric, the air flow entering the glovebox drops to 0. In this case $\cos \alpha = 1$.

$$\text{The } \Sigma_A V_r dA = \text{glovebox flow} = 16518 \text{ cc/sec}$$

$$\text{So } (e + Pv) \rho (\Sigma_A V_r \cos \alpha dA) = (e + Pv) \rho * 16518 \text{ cc/s}$$

$$\text{and } (e + Pv) \rho = 5773.84 \text{ J/s} / 16518 \text{ cc/sec} = 0.3495 \text{ J/cc}$$

$$\text{Then } (e + Pv) = 0.3495 \text{ J/cc} / 0.001191 \text{ g/cc} = 293.396 \text{ J/g}$$

and the pressure volume product $PV = 293.396 \text{ J/g} - 213.27 \text{ J/g} = 80.125 \text{ J/g} = 0.9416 \text{ L-atm/L}$

Therefore for a given Liter of gas the pressure must be 0.94 atmospheres

Using these calculations, Table 4 was prepared

Table 4- Glovebox 21-A Pressure

dQ/dt	$(d/dt \Sigma v_{epdV})$	$dW_{c.v.}/dt$	$\Sigma_A(e+Pv)\rho V_r \cos \alpha dA$		$(e+Pv)\rho$	$e+Pv$	Pv	P
J/s	J/s	J/s	J/s	cc/sec	J/cc	J/g	J/g	atm
10000	4226.1 6	0	5773.8 4	16518	0.3495 5	293.39 6	80.125 2	0.941
10050	4226.1 6	0	5823.8 4	16518	0.3525 8	295.93 6	82.665 9	0.971
10090	4226.1 6	0	5863.8 4	16518	0.3550 0	297.96 9	84.698 5	0.995
10100	4226.1 6	0	5873.8 4	16518	0.3556 0	298.47 7	85.206 6	1.001
10110	4226.1 6	0	5883.8 4	16518	0.3562 1	298.98 5	85.714 8	1.007

Table 3 shows that the glovebox 21-A air must be heated at 10100 J/sec is required to bring the glovebox atmosphere from -0.5 inches of water to 1 atmosphere.

A standard finite difference model was prepared wherein the surface of the reaction boat was assumed to be uniformly heated to the bounding temperature of 1561 K. This particular temperature was chosen as a reasonable bound to the expected hydride temperature in the reaction boats and also a temperature wherein the standard heat transfer mechanisms (conduction, convection, and radiation) dominate.

The finite difference model is included in Appendix A, and is briefly described here. The model begins at time 0 and initializes all of the properties of the gas. Examples of these properties are temperature, density, viscosity, heat capacity, and thermal conductivity. From these values and the temperature gradient between the reactive surface and the glovebox air, one may calculate the Prandtl and Grashof numbers. One may then calculate a convective heat transfer coefficient and calculate the heat transferred to the air convectively. Next the heat transferred by convection and by radiating directly to a metal shield arching over one side and above the powder boat was calculated. Then the heat transferred from the shield to the remainder of the glovebox was calculated via convection and radiation. Finally the glovebox air heat inventory and temperature are calculated.

The model then advances to the next time increment and the gas parameters are recalculated and a new cycle of heat transfer is initiated. Thus one can follow the glovebox pressure and air temperature in very small time increments.

Exercising this model in 0.02 second increments with a powder surface temperature at 1561 K, leads to the conclusion that the energy flow to the glovebox air is limited to approximately 540 J/s whereas 10100 J/s are required to bring the glovebox to 1 atmosphere. This shows that the heating rate would have to be increased by a factor of 18 to remove the negative pressure from the glovebox.

Establishing Acceptable Glovebox Temperatures

The finite difference model also allows the calculation of glovebox steel and air temperature. The model shows that the steel temperature does not vary by 1 degree celsius and the air temperature only rises 7 degrees celsius above ambient.

Appendix A

Documentation for Excel Heat transfer model

This spread sheet uses Sheet 1 to perform the model calculations and Sheet 2 to store foundational parameters. Sheet 2 was compiled through the application of several models, and Sheet 1 only uses a subset of the data contained in Sheet 2.

Sheet 1 is further divided into a section contained between columns A-H which is used for preliminary calculations, and columns I-BG which contain the heat transfer calculations. The purpose of the model contained in columns I-BG is to calculate the temperature of critical glovebox components when exposed to a high temperature surface, and to calculate the total energy flow rate into the glovebox air.

Columns A-H- give background info and the calculations required to establish the energy flow rate required to bring the pressure differential between glovebox 21-A and the atmosphere to 0. An atmospheric flow rate of 35 CFM and ambient starting temperatures are assumed for the calculation. The results of the calculations in columns A-H, are not necessary for temperature calculations of glovebox components as performed in columns I-BG.

Column I- the total time lapse in seconds. Initialized at 0

Column J- An arbitrary temperature in Kelvin units assigned to the heated surface. Initialized at 1561 K

Column K- the density of the glovebox air. Initialized at 0.001191 g/cm³

Column L- Heat capacity of the glovebox air. Initialized at 1.1026 J/K-g

Column M- Viscosity of the glovebox air. Initialized at 0.00018617 g/cm-sec.

Column N- The kinematic viscosity

Column O- The thermal conductivity of the glovebox air. Initialized at 0.00026653 J/sec-cm-K

Column P- The Prandtl number. Initialized at 0.77018

Column Q- The gaseous coefficient of expansion = 1/K Initialized at 0.003354

Column R- a collection of factors used in calculating the Grashof number

Column S- A characteristic length used for assessing convective heat transfer. Initialized at 18.36 cm

Column T- The temperature difference mathematically formulated by calculating the mean temperature of the heat source and the heat sink and subtracting the heat sink temperature. Initialized at 1262.8 K

Column U- The Grashof number. Initialized at 1.05E09

Column V- The convective heat constant = $k/L * 0.14 * (Gr * Pr)^{0.33} = 0.0018827 \text{ J/cm}^2/\text{sec/K}$

Column W- Area of the heated source. Initialized at 400 cm²

Column X- The heat in joules directly from the heated surface to the glovebox air. Initialized at 0 J

Column Y- Blank- used as visual spacer

Column Z- Emissivity of heated surface. Initialized at 0.7

Column AA- Radiation heat transfer constant. A value given for Pu dioxide Initialized at 5.4538 J/sec/cm/R⁴

Column AB- Heat in Joules transferred radiatively from the heated surface to the overhanging airlock. Radiative heat transfer to the airlock is assumed to occur with 100% efficiency even though the airlock steel does not completely surround the combustion boat. The degree of coverage is sufficient to minimize the error. Calculation of shape factors is a potential refinement of the model. Initialized at 0

Column AC- The air inventory in g-moles within the airlock. Initialized at 1.35 g-mole

Column AD- The air temperature within the airlock. Initialized at 298.15 K

Column AE- g-moles of gas transferred convectively out of the airlock to the glovebox. Initialized at 0 g-moles

Column AF- An inventory of the heat contained within the air within the airlock. Initialized at 1.28E04 J

Column AG- Joules of energy convected out of the airlock. Initialized at 0 J.

Column AH- Joules of energy convected into the airlock. Initialized at 0 J.

Column AI- Joules of energy transferred convectively to the steel in the airlock from the airlock air. Initialized at 0 J.

Column AJ- Heat capacity of the airlock air. Initialized at 31.8 J/g-m/K

Column AK- Mass of steel in Airlock in grams. Initialized at 5750 grams

Column AL- The bounding temperature change of the steel in the airlock, ignoring conductance to the glovebox. Initialized at 0 K

Column AM- The bounding airlock steel temperature. Initialized at 298.15 K

Column AN- The surface area of the airlock presented to the glovebox air. Initialized at 2300 cm²

Column AO- Heat transferred convectively from airlock steel to glovebox air. Initialized at 0 J.

Column AP- Sum of heat transferred convectively from airlock air and airlock steel to glovebox air. Initialized at 0 J/sec.

Column AQ- Heat transfer rate to glovebox air necessary to bring pressure dP to 0. Initialized at 10095 J/sec

Column AR- Blank, used as visual spacer

Column AS- g-moles of glovebox air. Initialized at 124.22 g-moles

Column AT- Inventory of heat in Joules for glovebox air. Initialized at 1176074 J.

Column AU- Blank, used as visual spacer

Column AV- g-moles of glovebox air removed per time interval. Initialized at 0 g-moles

Column AW- Joules of energy removed from glovebox due to air flow out of glovebox. Initialized at 0 J.

Column AX- Joules of energy added to glovebox due to air flow into glovebox. Initialized at 0 J.

Column AY- Energy differential caused by air flow thru glovebox. Initialized at 0 J.

Column AZ- Blank, used as visual spacer

Column BA- Joules of energy absorbed by glovebox air. Initialized at 0 J.

Column BB- Inventory of energy in Joules within the glovebox steel. Initialized at 5.01E07 J.

Column BC- Glovebox temperature. Initialized at 298.15 K

Column BD- Joules of energy transferred convectively to glovebox. Initialized at 0 J.

Column BE- Temperature differential for glovebox. Initialized at 0 K.

Column BF- Heat capacity of glovebox air. Initialized at 31.7553 J/g-m/K

Column BG- Glovebox air temperature. Initialized at 298.15 K

Appendix C

Letter, T. D. Cooper, BWHC, to E. P. Bonadie, BWHC, "Scientific Evaluation of Safety in Processing Pu Metal", 15F00-99-054, dated May 25, 1999.

B & W Hanford Co.

a McDermott company

To	E.P. Bonadie	T5-55	
From	T.D. Cooper	T5-12	File No. or Ref.: 15F00-99-054
Subj	SCIENTIFIC EVALUATION OF SAFETY IN PROCESSING PU METAL		Date: May 25, 1999

References

- 1 HNF-2061 Gas Generation from Plastics during Vault Storage by Way of Radiolysis Reactions
- 2 HNF-2062 Gas Generation from Plastics during Vault Storage by Way of Thermolytic and Oxidation Reactions
- 3 Letter # 15F00-99-045 "Calculations Bounding Glovebox HA-21A Pressure and Temperature" Rev.1
- 4 Haschke JM et.al. 1983, "The Reaction of Plutonium with Water, Kinetic and Equilibrium Behavior of Binary and Ternary Phases in the Pu + O + H System", RFP-3416
- 5 Haschke JM et.al. 1997a, "Oxidation Kinetics of Plutonium in Air from 500 to 3500 °C: Application to Source Terms for Disposal", La-UR-97-360
- 6 Pichelin G. and Rouanet A., 1991, "Predictive Modeling of High-Temperature Chemical System Vaporization under Atmospheric Pressure", Chemical Engineering Science, Vol 46, No. 7 pp 1635-1649

1) Introduction

The Plutonium Finishing Plant at Hanford has stored plutonium metal ingots in vault storage for times varying from approximately 12 years to 30 years. Many different storage package configurations were used, although the most common initial types were:

- * metal ingot- plastic bag- slip lid can sealed with plastic tape
- * metal ingot- food pack can- plastic bag- food pack can

The plastic bag used at PFP for metal storage was PVC before the late 1970's. In the late 1970's it was realized that PVC deteriorated quickly under vault storage conditions and the contamination control bag was changed to polyethylene. This change did not affect most of our metal inventory, since the majority of our Pu metal has not been re-canned since polyethylene came into use.

In the late 1970's, it was decided to overpack all of the metal cans with additional food pack cans. From 1 to 2 overpack cans were used to further slow the diffusion of atmospheric gases to the metal. All of the cans were not weighed in the intermediate storage configuration, and therefore weighing the finished configuration does not allow calculation of the fixation rate of atmospheric gases upon the metal.

Most of the Pu metal in the vault has hardly been disturbed for approximately 20 years except for the fact that approximately 63 have been radiographed. The 63 radiographed cans show varying quantities of corrosion products ranging from approximately 5 grams to perhaps 200 grams of powder. These corrosion products must result from the reaction of Pu metal with gases that were either initially present in the inner container, were generated during storage, or that leaked in from the atmosphere.

The composition of the corrosion product is not known. Potential reaction products of gases with Pu metal are plutonium oxides, plutonium hydride, plutonium nitride, and plutonium carbonate. For very leaky seals, the corrosion product can be described as primarily plutonium dioxide with small inclusions of hydride, nitride, or metal. For storage configurations with an excellent inner can seal, the corrosion products would be expected to be primarily plutonium sesquioxide, plutonium hydride, and plutonium nitride.

In the latter case, it would not be unusual for the Pu metal to absorb all or most of the inner can atmosphere causing a vacuum within the inner can. The inner vacuum may cause the inner can to collapse or "panel" thereby causing an inward deflection of the can walls.

Upon opening a paneled can in 1996, sparks and apparent flames about the size of a person's hand were seen and photographed to emanate from a paneled inner can. The flame above the can was visible for approximately 80 milliseconds. After opening the can, and dumping the metal ingot and associated corrosion products into a metal boat, the corrosion powder glowed red and the metal ingot showed reactive areas that ignited and glowed for several minutes.

In evaluating this event, it was realized that this was the most reactivity anyone could remember seeing upon opening a stored metal item. The reactive corrosion product identities, masses, concentrations, and reaction rates were not known.

In an attempt to provide bounding estimates, it was postulated that all of the hydrogen on the plastic bag could be released through radiolysis as either hydrogen gas, hydrochloric acid (HCl), or through thermal oxidation of the hydrogen to water. It was calculated that transferring all of the hydrogen to the metal could form 150 grams of plutonium hydride.

A bound for the reaction rate was the next goal. If one assumes all of the reaction heat (from oxidizing 150 grams of plutonium hydride) is transmitted to glovebox HA-21A air, one may calculate a pressure rise of 10.5 psig. This pressure rise is undoubtedly high because it is unrealistic to think that all of the reaction heat is deposited in the air. Due to the high temperature reached in a cloud flame, radiant heat transfer would effectively transfer a good share of the heat to the glove box walls.

From other combustion experiments, it was deduced that burning 150 grams of uranium hydride as a cloud in glovebox HA-21A gives a pressure increase of 4.5 psig. An instantaneous pressure increase of 3 psig is capable of blowing gloves off of a glovebox and is not an acceptable process

condition. Since these bounding calculations give unacceptable answers, it is necessary to further bound the total energy releasable, or the energy emission rate.

2) Energy Release Rates

Energy release rates for a given reaction can be moderated by such factors as reactant concentrations, temperature, or constraints upon the movement of reactants. Since we are dealing with atmospheric gases igniting pyrophoric powders, we cannot reduce the gas concentrations, and we cannot reliably control the temperature. We must instead rely upon gaseous diffusion control.

Three diffusion control mechanisms are operable. The first is the diffusion of oxygen through nitrogen to reach the powder surface. The second is the diffusion of all gases through the powder bed matrix to reach reactive surfaces within the bed. The third is diffusion of gases into and throughout a given particle. Since we need further data to model the last two diffusion mechanisms and since it is estimated that several years and several millions of dollars would be required, we shall invoke only the first diffusion control mechanism quantitatively to provide further reaction limits.

The reaction rates for PuH_2 with oxygen were shown by Stakebake to be a function of temperature up to 360 °C. Above 360 °C, its reactivity is difficult to correlate to temperature since the sample temperature ceases to follow the furnace temperature and tends to heat itself autocatalytically up to a diffusion controlled limit. Above 500 °C, (see Appendix A) Haschke showed that Pu metal oxidation becomes diffusion controlled. Since plutonium hydride is very reactive and its specific surface area is approximately 20,000 times higher than for Pu metal, one might expect that the reaction rate of PuH_2 will transition from being Arrhenius to diffusion controlled at temperatures between 360 and 500 °C.

The diffusion-controlled oxygen concentration gradient from Haschke's experiment was used to calculate the diffusion-controlled reaction rate of hydride using in this case a nominal specific surface area of 2000 cm^2/g of plutonium hydride. Three conditions, plutonium hydride in a cloud, plutonium hydride in the inner can, and plutonium hydride spread evenly in a powder boat, were analyzed for the maximum oxygen delivery and reaction rates. The results of these calculations are shown in Table 1.

Table 1- Enthalpy Liberated in Exposing PuH_2 to Air

Hydride Configuration	Limiting Mechanism	O ₂ flow g-m/sec	Molar dH J/g-m O ₂	dH J/sec
Powder cloud	diffusion	6.92	801533	5546611
Powder inner can	convection	0.00382		3062
Powder boat	convection	0.037		29657

In making the above calculations, several assumptions were necessary. A bounding air temperature of 2000 °C was assumed. For the powder cloud case, it was assumed that oxygen is absorbed at the diffusion-controlled limit. For the powder in the inner can and in the powder boat, oxygen delivery rates were calculated using diffusion and convection models.

Areas needing further clarification are:

- * What energy release rates are permissible in Glovebox HA-21A?
- * What effect does intra-bed diffusional limitations have upon the reaction rates?
- * What conditions are required to transform a hydride powder bed into a cloud? Are mechanisms available for forming clouds under process conditions? How fast does hydride react under spilling or pouring conditions and are these conditions equivalent to combusting hydride as a cloud?

The first effort to limit the energy of the system was to calculate the total amount of hydrogen that could be transferred to the Pu metal from both radiolysis and thermal-oxidative mechanisms. The results of these studies are contained respectively in reference 1 (HNF-2061) and reference 2 (HNF-2062). The results of these studies show that conservatively, all of the hydrogen on the bags can transfer to the metal thus forming 150 grams of hydride. In the case of very slowly leaking inner cans, even larger amounts of nitride and sesquioxide can form.

Reference 3 contains the analysis showing the limiting energy release for glovebox HA-21A. A limit of 12,600 J/sec is sufficient to raise the glovebox pressure from -0.5 inches of water to 0 inches of differential pressure. The concurrent glovebox air temperature rise is 7 °C.

Erasing the negative pressure differential does not massively endanger glovebox containment, but is a prerequisite condition that can lead to contamination seeping through seals.

In comparing the enthalpy production rates in Table 1 with the 12,600 J/sec limit derived in reference 3, one concludes:

- * Combustion of a hydride cloud is definitely unacceptable
- * Combustion of hydride within an inner can is acceptable as long as the powder remains in a bed within the can
- * Combustion within a powder boat is not acceptable without invoking further diffusional limitations.

3) Issues Not Resolved Mathematically

Mathematically modeling intra-bed and intra-particle diffusion processes and modeling the creation of clouds from powder beds, is estimated to require several years and millions of dollars in expense in measuring the required data. It was decided instead to rely on the practical experiences of several sites that have handled stored metal and hydride powders. To this end, telephone conversations were held with experts from Rocky Flats, Lawrence Livermore National Lab, and Los Alamos National Lab. In addition a workshop was convened at Denver in November 1998 for face to face meetings with recognized plutonium handling experts from Savannah River Site, Los Alamos National Labs, Lawrence Livermore National Labs, Rocky Flats Environmental Transition Site, Idaho Falls, and Hanford. This meeting was also attended by DOE Headquarters representatives and the National Defense Safety Board representatives.

The conclusions reached from all of the telephone conversations and the Denver Workshop were that no one in the entire DOE complex has ever reported overheating or pressurization from burning single ingots of plutonium metal or handling up to 200 grams of plutonium hydride in an air filled glovebox. Hydride has been handled and burned in a bed configuration and has also been poured from crucible to crucible without causing cloud formation.

This process history is strong evidence that intra-bed diffusional limitations are very important in controlling the energy production rate. We have not quantified this phenomena, but consider the practical experience to be sufficient to close the investigation. This same process history also shows that accidental cloud formation of hydrides has never been observed and is therefore an extremely unlikely event. It is common to see a tiny fraction of the powder (believed to be sub-micron particles) lift into the air and sparkle, but no one has ever reported lifting of the majority of the powder into a cloud except in carefully controlled conditions designed to accomplish that goal.

We therefore believe that mathematical proof has been developed that opening and reacting Pu corrosion products within the inner storage container in air is acceptable. The practical experience base says that reacting up to 200 grams of plutonium hydride within a powder boat is also acceptable and that accidental cloud formation involving the majority of the hydride has not historically occurred within the DOE complex, and should be considered a very improbable event.

If more than 200 grams of plutonium corrosion products were discovered in a single container within the Hanford vault, it is believed that unreactive plutonium dioxide would be the most common corrosion product. In the rare case involving a very-very slowly leaking inner container wherein large amounts of nitride and sesquioxide could possibly build, reaction rates will still be safely controlled by oxygen convection rates through vessel openings and intra-bed and intra-particle oxygen diffusion limitations.

It is considered more probable that high percentages of hydrides could accumulate in small batches of corrosion products than large batches. Since no experience base exists for handling more than 200 grams of pure plutonium hydride in an air-filled glovebox, it is considered prudent to limit any planned accumulations of plutonium metal corrosion products from multiple storage containers to 200 grams.

From all of these considerations I have concluded that the contents of a single Pu metal ingot storage container at Hanford are not expected to challenge the pressure or temperature limits for Glovebox HA-21A.

Appendix A- Diffusion Controlled Reaction Rates of Pu in Dry Air

The reaction rates of unalloyed non-hydrided Pu metal with dry air over the temperature range ambient to 500 °C (Haschke 1983) are given in Figure A1 as a function of reciprocal absolute temperatures.

Figure A1 - Reaction Rates of Unalloyed Pu with Dry Air

between 500 °C and 925 °C, the reaction rate does not change appreciably for static Pu metal, as seen in Table A1 (Haschke 1997a)

Table A1 - Pu Metal Oxidation at Combustion Temperatures

Temp. °C	Metal Type	Pu Mass Reacted (g)	Initial Area cm ²	Reaction Period min.	Oxidation Rate gPu/cm ² /min
500 ±60	unalloyed	202	34	60±20	0.1±0.07
775±50	alloyed	997	140	75±5	0.1±0.07
860±40	unalloyed	89	34	23±7	0.11±0.04
900±50	unalloyed	570	68	40±5	0.21±0.09
925±50	unalloyed	1770	188	55±5	0.17±0.08

Table A1 shows that the unalloyed non-hydrided Pu metal exhibits a plateau in reactivity between 500 and 860 °C, with only modest increases at 925 °C.

This is clear evidence that the reaction has ceased to be Arrhenius controlled, and has become diffusion controlled. One may calculate the oxygen diffusion gradient using Fick's law:

$$dw/dt = -D \cdot A \cdot dc/dx$$

where:

dw/dt is the oxygen flux in moles $O_2/cm^2/sec$

D is the diffusivity coefficient in cm^2/sec

A is the diffusional cross sectional area = 1 cm^2

dc/dx is the concentration gradient in g-moles $O_2/cm^3/cm$

dw/dt can be calculated from the Pu reaction rate

D the diffusion coefficient is temperature dependent and is calculated from the Chapman-Enskog relationship (Pichelin, 1991) :

$$D_{ib} = 0.001853 \cdot \text{Sqrt}(T^3 \cdot (1/M_i + 1/M_b)) / (P \cdot \text{Sigma}_{ib}^2 \cdot \text{Omega}_{ib})$$

where

D_{ib} is the diffusion coefficient of gas i through gas b

T is the absolute temperature

M is the molecular weight

P is the total gas pressure

Sigma is the collision diameter

Omega is collision integral

A is set equal to 1 cm^2

So that dc/dx can be calculated as $(dw/dt)/(D \cdot A)$

The results of this calculation are presented in Table A2

Table A2 - Calculation of Oxygen Diffusion Gradients

Temp °C	gPu/cm ² /min	g-moles O ₂ /cm ² /sec	diffusion coefficient	Oxygen Gradient g- moles/cm ³ /cm
25	2.2E-10	1.53E-14	0.178	8.62E-14
100	1.35E-07	9.45E-12	0.249	3.79E-11
200	2.99E-05	2.08E-09	0.356	5.85E-09
300	1.00E-03	6.99E-08	0.474	1.47E-07
400	1.18E-02	8.26E-07	0.603	1.37E-06
500	7.39E-02	5.15E-06	0.743	6.93E-06
600	1.0E-01	6.97E-06	0.892	7.82E-06
673	1.0E-01	7.67E-06	1.01	7.62E-06
775	1.0E-01	6.97E-06	1.17	5.94E-06
860	1.1E-01	7.67E-06	1.32	5.82E-06
900	2.1E-01	1.46E-05	1.389	1.05E-05
925	1.7E-01	1.19E-05	1.43	8.27E-06

The oxygen gradient is graphically compared to the temperature in Figure A2. The gradient is not smooth at higher temperatures because of the relatively large measurement errors, however it is apparent that the gradient has plateaued at about 500 °C, and that the reaction has become diffusion limited.

Figure 2 - Oxygen Gradients at the Surface of Reacting Unalloyed Pu Metal

Since Figure A2 shows that the combustion of unalloyed non-hydrided Pu metal becomes diffusionally limited beyond 500 °C, it is reasonable to believe that very reactive compounds with high surface area should also become diffusionally limited beyond 500 °C. This principle will be used to estimate the reaction rate and energy production rate of a non-hydrided metal ingot; a hydrided ingot, 150 grams of Pu hydride within an inner can, 150 grams of Pu hydride within a powder boat, and 150 grams of Pu hydride within a dust cloud.

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