



## Department of Energy

Washington, DC 20585

FEB 16 2000

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DNF SAFETY BOARD

The Honorable John T. Conway  
Chairman  
Defense Nuclear Facilities Safety Board  
625 Indiana Avenue, N.W.  
Suite 700  
Washington, D.C. 20004-2901

Dear Mr. Chairman:

This letter informs you of the completion of Commitment 113, "Document a Decision for Polycubes Stabilization Path Forward," included in the Implementation Plan (IP) for the Defense Nuclear Facilities Safety Board (DNFSB), Recommendation 94-1, Remediation of Nuclear Materials in the Defense Nuclear Facilities Complex, Rev. 2. The enclosed documentation, *White Paper Supporting Decision to Stabilize Polycubes using Direct Oxidation in Muffle Furnaces*, provides supporting preliminary technical details related to the above-mentioned decision.

The selected path forward for polycube stabilization at the Plutonium Finishing Plant is to use a one-step muffle furnace process instead of a two-step process, i.e., pyrolysis followed by muffle furnace stabilization. This path forward will provide significant opportunity for cost reduction and schedule recovery. Additional testing is required to develop processing parameters necessary to determine estimated throughputs within acceptable limits. After integration with the balance of plant activities, a schedule will be developed by February 29, 2000, to support the IP commitment to determine a completion date for polycube stabilization.

We continue to closely track progress on all Recommendation 94-1 commitments and will keep you and your staff apprised of our progress. If you have any questions, please contact me at (202) 586-5151 or Mark W. Frei, Deputy Assistant Secretary for Project Completion, at (202) 586-0370.

Sincerely,

David G. Huizenga  
Deputy Assistant Secretary  
for Integration and Disposition  
Office of Environmental Management

Enclosure

cc w/enclosure:  
M. Whitaker, S-3.1



RL-F-1325.6 (02/00)

United States Government

Department of Energy  
Richland Operations Office**memorandum**

DATE: FEB 7 2000  
REPLY TO  
ATTN OF: MDD:MRH/00-MDD-036

SUBJECT: PLUTONIUM FINISHING PLANT (PFP) COMPLETION OF DEFENSE NUCLEAR FACILITIES SAFETY BOARD 94-1 IMPLEMENTATION PLAN, REV. 2, COMMITMENT 113, "DOCUMENT A DECISION FOR POLYCUBES STABILIZATION PATH FORWARD"

TO: David G. Huizenga  
Deputy Assistant Secretary  
for Integration and Disposition,  
EM-20, HQ

- References:
- (1) FDH letter, A. Clark, J. L. Buelt, L. J. Olguin to P. M. Knollmeyer, "White Paper Supporting Decision to Stabilize Polycubes Using Direct Oxidation in Muffle Furnaces" (9959367), dated December 29, 1999.
  - (2) RL memorandum, K. A. Klein to C. L. Huntoon, EM-1, "Proposed Revision to the Plutonium Finishing Plant Portion of the Defense Nuclear Facilities Safety Board Recommendation 94-1 Implementation Plan" (00-MDD-016), dated November 8, 1999.

This memorandum reports completion of the Implementation Plan (IP) for the Defense Nuclear Facilities Safety Board (DNFSB), Recommendation 94-1, Remediation of Nuclear Materials in the Defense Nuclear Facilities Complex, Rev. 2, Commitment 113, "Document a Decision for Polycubes Stabilization Path Forward," due date for the commitment was January 2000. Reference 2 provided a proposed revision to the PFP portion of the DNFSB Recommendation 94-1 IP which includes adding the subject commitment. Actual completion date for the commitment was December 29, 1999.

A one-step muffle furnace process will be employed to stabilize polycubes (Reference 1). This approach will provide significant opportunity for cost reduction and schedule recovery. Additional testing is required to develop processing parameters necessary to determine estimated throughputs. After integration with the balance of plant activities, a schedule will be developed to support the proposed IP Commitment to determine a completion date for polycube stabilization by February 2000.

David G. Huizenga  
00-MDD-036

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FEB 7 2000

The stabilization process utilizing the pyrolysis equipment developed by Los Alamos National Laboratories (LANL) will be retained as a backup alternative. The equipment has been crated up and may be shipped to Hanford for storage.

If you have any questions, please contact me, or your staff may contact Peter M. Knollmeyer, Assistant Manager for Nuclear Materials and Facility Stabilization, on (509) 376-7435.



Keith A. Klein  
Manager

cc: C. J. Sink, Jr., EM-21  
M. W. Frei, EM-40  
R. L. Kaltrcider, EM-43  
J. E. Newson, EM-43  
J. A. Turi, EM-43  
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L. J. Olguin, FHI  
A. F. Shattuck, FDNW  
G. W. Jackson, WSMS

**FLUOR DANIEL**

Fluor Daniel Hanford Inc  
P O Box 1000  
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December 29 1999

FH-9959367

Mr P M Knollmeyer Assistant Manager  
Nuclear Materials and Facility Stabilization  
U S Department of Energy  
Richland Operations Office  
P O Box 550  
Richland Washington 99352

Dear Mr Knollmeyer

**WHITE PAPER SUPPORTING DECISION TO STABILIZE POLYCUBES USING  
DIRECT OXIDATION IN MUFFLE FURNACES**

Reference Memo S A Jones FH to R K Leugemors FH same subject  
15F00-99 16S dated December 29 1999

Fluor Hanford has reviewed the option for processing the Plutonium Finishing Plant (PFP) inventory of Plutonium (Pu) polycubes by thermal stabilization (muffle furnace) without prior pyrolysis processing. We have determined that the current or similar muffle furnaces in use at PFP can and will be utilized to safely and more efficiently stabilize these items.

This decision is based on a technical evaluation performed by the Plutonium Plant Support Laboratories (PPSL) as documented in the above Reference which incorporates the results of testing and analysis completed to date by PPSL and the Pacific Northwest National Laboratories. This report documents that polycube processing can be performed at a 400 gram loading and still maintain the peak flammable off gas below 25 percent of the lower flammability limit. This loading combined with conservative process cycle periods indicates that thermal stabilization can be performed more efficiently than pyrolysis. This will also eliminate the expenditure for new equipment installation and will support an earlier startup of polycube stabilization.

A Kepner Trego evaluation was performed on December 22 1999 by a diverse group of technical and project personnel. The review included a comparison of direct thermal processing vs pyrolysis followed by thermal processing in the following areas: safety, ability to meet 3013 and Materials Disposition Standards, ALARA, technical confidence, schedule, cost, facility logistics, process reliability, and maintainability. The



Mr P M Knollmeyer  
December 29 1999  
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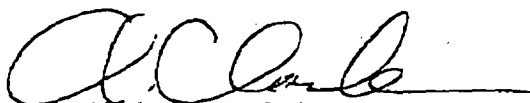
FH-9959367

results showed that direct thermal stabilization was clearly preferential to pyrolysis followed by thermal stabilization especially in the categories of safety, schedule and cost

This letter closes the interim commitment to identify the polycube stabilization process as identified in the anticipated December 1999 update of the Defense Nuclear Facility Safety Board (DNFSB) 94-1 Implementation Plan. A revised schedule for completion of PFP polycube stabilization will be developed based on this selected path forward for polycube processing the results of subsequent polycube testing at PFP and PNNL and integration of the revised path forward with other DNFSB 94-1 commitments

If you have any questions or require additional information please contact Mr R K Leugemors at 373 0729

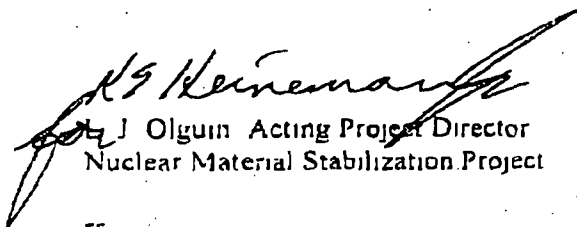
Sincerely



A Clark Senior Director  
Project Planning and Integration  
Nuclear Material Stabilization Project



J. L. Buelt Product Line Manager  
Process and Measurement Technology  
Pacific Northwest National Laboratory



J. Olguin Acting Project Director  
Nuclear Material Stabilization Project

rs

Attachment



ATTACHMENT  
FH-9959367

## FLUOR DANIEL HANFORD, INC

### INTEROFFICE CORRESPONDENCE

To R K Leugemors T2 12 Date 15F00 99 168  
December 29, 1999

From S A Jones *S Jones* T5-12 Telephone 373 3347

cc Distribution

Subject WHITE PAPER SUPPORTING DECISION TO STABILIZE POLYCUBES USING DIRECT OXIDATION IN MUFFLE FURNACES

- References (1) Lewis, W S, "Stabilization of Polycubes Engineering Study", WHC SD CP-TI-204, September 1996, Westinghouse Hanford, Company, Richland, WA
- (2) Jones, S A, G S Barney, and J Abrefah "Muffle Furnace Stabilization of Polycubes", September 9, 1999 B&W Hanford Company Richland, WA

#### Purpose

As part of stabilizing the inventory of plutonium at the Plutonium Finishing Plant, polycubes must be treated to produce plutonium and uranium oxides in a form suitable for storage. This white paper will provide the technical justifications for the decision to stabilize polycubes using muffle furnace oxidation. Detailed technical reports will be issued separately for the laboratory studies and calculations that have been performed to support this decision.

#### Background

##### Origin of Polycubes

Nuclear criticality studies at Hanford in the 1960's and 1970's used polycubes in order to prepare items with fixed H/X ratios. The ratio of hydrogen to fissile atoms affects neutron moderation. Using these solid forms, simulated "solutions" that were easy to handle and contained high concentrations of fissile material could be prepared. Polystyrene powder was mixed with plutonium and/or uranium oxides and heat pressed into "cubes". The dimensions were 2 inches wide by 2 inches deep with heights of 1/2, 1, 1 1/2, or 2 inches. The concentration of plutonium was such that H/X ratios of 5, 15, or 50 were achieved. Most of the polycubes remaining in the Hanford inventory had an H/X ratio of 50 before radiolytic decomposition.

##### Polystyrene Thermal Degradation

When polystyrene is heated in an inert environment, pyrolysis of polystyrene produces styrene monomer, toluene, ethyl benzene, and other alkyl benzene compounds. In the inert environment,

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all of the polystyrene is converted to hydrocarbons and a carbon char. Destruction of the char requires heating in an oxidizing atmosphere.

When heated in air, pyrolysis also occurs to produce styrene, toluene, and ethyl benzene but some of the polystyrene is oxidized producing water, carbon monoxide, and carbon dioxide. The thermal decomposition of polystyrene in air, generating these gaseous products is strongly influenced by temperature and the temperature ramp rate. A change of temperature not only causes the reaction rate to change but also changes the mechanism. Oxidation of polystyrene begins at a lower temperature than pyrolysis.

#### Historical Treatment

Up until about 1985, the plutonium in polycubes was recovered in the Plutonium Reclamation Facility (PRF). From the inventory of material at the Plutonium Finishing Plant it appears the highest plutonium concentration cubes were processed. What remains today are approximately 1600 polycubes containing about 35 kg of plutonium. Most of these polycubes appear to be of similar composition. This conclusion was reached by examining the item identification numbers, the plutonium content of the individual items, and the age of the items.

The first method to recover plutonium from polycubes involved burning polycubes in the Plutonium Reclamation Facility. Any tape and paint coating was removed from the polycubes. Using a hammer and cold chisel the polycubes were broken up into smaller pieces and placed into a Hoskins type furnace at 600 °C where they burned. During heating, air was pulled through the furnace and four wires maintained at red heat above the crucible ignited vapors. Decarbonization was considered complete when there was no longer any flame or glowing in the crucible. This process produced plutonium oxide of acceptable quality. However, this process produced considerable amounts of soot that plugged off gas filters. The temperature was difficult to control once the ignition temperature of the styrene monomer was reached. Controlling the temperature was significant in this time, because the higher fired oxides were more difficult to dissolve. This plutonium recovery was being performed during a time when plutonium weapons production was still active and the quality of the oxide for this purpose was an important consideration.

A new process developed in 1969 consisted of removing the coatings, distilling styrene monomer in an inert environment, and decarbonizing the oxide product of the distillation. This method was discontinued due to many problems including inadequate collection of the styrene monomer in a condenser. During this operation, styrene monomer was observed outside the distillation apparatus condensing and running down the interior glovebox walls.

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Reason for Stabilizing Polycubes

The current storage configuration for polycubes is unacceptable for long term storage. Because radiolytic decomposition of polystyrene by the decay plutonium generated gases that pressurized sealed containers, these items are stored in a vented configuration. Radiolysis has degraded the polystyrene. When the polycubes were first placed in these storage containers they could be handled with little concern for contamination control because a coating of paint and tape sealed the polystyrene and plutonium cubes. Containers were vented and a filter placed over the opening so that gases produced by radiolysis could escape while maintaining contamination control. By 1985, the polystyrene had degraded enough that in handling the containers and inverting them, an air borne contamination was produced. Decomposition of the glue holding the filter in place allowed radioactive particles to escape from the storage container during inversion of the storage container. The filters glued over the opening are adequate for storage when the items are not moved.

Effect of Radiation on Polystyrene

Pure polystyrene and the polystyrene in actual polycubes differ because of the effect of radiation on the polystyrene. High energy radiation is nonspecific and all chemical bonds have a certain probability of involvement, which depends on their electron density. The process rarely consists only of chain scission, usually cross linking occurs simultaneously. One effect of radiation on polystyrene is to cause it to lose hydrogen. The initial effect is to produce cross linking. Cross linking of the polystyrene in polycubes has lowered their mechanical strength and has caused the polystyrene to have a higher pyrolysis temperature.

Radiation damage to polystyrene increases radical production, which increases the rate of polystyrene oxidation. Polymer oxidation reactions generally show a slow induction period during which no visible changes occur in the material. Oxidation leads to decreased molecular weight and discoloration of the polymer. In the presence of air, auto oxidation also occurs through reactions with atmospheric oxygen. These polycubes were stored in vented containers to prevent pressurization from the release of hydrogen. This means that oxygen was present. Evidence of oxidation is seen in that the polycubes have become friable.

A report of results of critical experiments performed with polycubes having an H/Pu ratio of 50 listed the isotopic abundances of the plutonium isotopes that had a  $^{240}\text{Pu}$  isotopic content of 18.35% in 1970. Calculating the decay of  $^{241}\text{Pu}$  and the in growth of  $^{241}\text{Am}$ , these polycubes now contain approximately 5%  $^{241}\text{Am}$  relative to total plutonium. This americium concentration is consistent with the high radiation doses reported on the containers of polycubes.



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Calculations based on the amount of plutonium in the polycubes in contact with polystyrene for 30 years yield that the polycubes would have been exposed to a dose of 10 to 100 megarads during their storage. This dose is sufficient to have reduced the hydrogen content in the polycubes significantly. By comparison, freshly prepared polyethylene polycubes with an H/X ratio of 15 decreased to an H/X ratio of 14.28 in 98 days. Based on the 30 years since the preparation of these polycubes, the amount of hydrogen in the system is significantly lower than was originally present.

As a result of radiolysis, the amount of hydrogen containing carbon compounds is lower in irradiated polycubes compared to pure polystyrene. Consequently, the flammable organic compounds generated in thermal stabilization of degraded polycubes will be lower compared to that observed in pure polystyrene.

Historical Baseline Using Pyrolysis

The current baseline process was selected based on an engineering study of polycube stabilization that concluded the pyrolysis method developed at Los Alamos National Laboratory (LANL) with a silent gas plasma off gas treatment was the preferred alternative (Lewis, 1996). The off gas treatment was later changed to a catalytic oxidation process. The pyrolysis method involves a device that heats the polycubes to 750 °C in an inert atmosphere. Flammable organic products of the pyrolysis are collected in a reservoir. After accumulating sufficient quantity of these flammable compounds, a series of pumps and valves feed the compounds and air into a catalytic converter that oxidizes the hydrocarbons. Monitors for hydrocarbons provide controls for the process through a computer feedback system. Although considerable effort has been invested at LANL into the design and optimization of this process using pure polystyrene, no tests were conducted using actual polycubes. The residue from the pyrolysis step must be transferred to a muffle furnace where the carbon char is oxidized to complete the stabilization process. This stabilization process was identified as the baseline in the 1999 submittal of the PFP Integrated Project Management Plan (IPMP).

Proposed Baseline Change to Direct Oxidation

Studies of the gases generated when polystyrene was heated in air were performed in the PFP Plutonium Process Support Laboratories (PPSL) as part of the plutonium sludge stabilization safety analysis. It was found that when polystyrene was heated in flowing air, a significant portion of the polystyrene was directly oxidized without combustion. Similar results were identified during initial tests at PNNL conducted for evaluation of the LANL pyrolysis processes. The results of these studies indicated that it might be possible to safely stabilize polycubes using the existing muffle furnaces at PFP. Additional studies were performed in the PPSL and at Pacific Northwest National Laboratory (PNNL). The results of these studies are the

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basis of the decision that muffle furnace oxidation should be used rather than pyrolysis for the stabilization of polycubes

Description of Current Muffle Furnace Operations

The muffle furnaces currently used for stabilization of plutonium oxides scraps, and sludges have an internal furnace volume of approximately six liters. A programmable temperature controller is used to adjust the temperature ramp rates including dwell times at fixed temperatures. Air is drawn in through the space around the loosely fitted door of the furnace using a vacuum source at the back of the furnace separate from the glovebox ventilation. For some feed material carbon dioxide is added to the air in the furnace to reduce the potential for a flammable gas mixture developing in the furnace. Feed material is placed in stainless steel boats and then inserted into the furnace. After the heating cycle is completed the furnace is allowed to cool before the boat is removed.

**Direct Oxidation Process**

An accumulation of organic vapors above the lower flammability limit inside a plutonium handling glovebox is an unacceptable situation. LANL pyrolysis equipment uses an argon atmosphere to exclude oxygen during the pyrolysis. The hydrocarbons generated by that process are oxidized in a separate step using a metering pump and valves to control the amount of hydrocarbons and air combined at any time.

The proposed process for direct oxidation of polycubes in muffle furnaces will produce flammable gases in the air at very low concentrations. Tests were designed to measure the rate of flammable gas generation and the composition of the gases resulting from direct oxidation of pure polystyrene, simulants and actual polycube samples.

Testing at Hanford was conducted in three areas

- 1 At PPSL, G Scott Barney tested synthetic polycubes using polystyrene, polystyrene mixed with cerium oxide to simulate the effect of plutonium oxide, and polystyrene cross linked with divinyl benzene to simulate the effect of radiation on polycubes. These tests were performed on milligram scale tests in a Thermal Analyzer Mass Spectrometer (TA MS) system and on multigram samples in a muffle furnace.
- 2 At PNNL, a very similar TA MS system was used by John Abrefah to test milligram-sized samples of actual polycubes. These actual polycubes were selected as ones that were representative of the inventory of polycubes to be stabilized.
- 3 Also at PPSL, Susan Jones tested full sized, actual polycubes in a muffle furnace. These polycubes contained less radioactivity than the ones tested at PNNL. These are thought to be the worst case polycubes.

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A number of conservative assumptions have been made for the safety analysis. Pure polystyrene was used for calculation of the maximum gas generation rate. Experiments using varying degrees of cross linking in pure polystyrene have demonstrated that the gas generation rate decreases with increasing cross linking. Additionally, experiments with the actual polycubes at PNNL and PPSL show that the rate of weight loss is slower than with pure polystyrene. This has been demonstrated using small (up to 600 mg) samples in the TA-MS and using full sized polycubes (up to 218 g) in the laboratory muffle furnace.

The laboratory muffle furnaces are much smaller than the units used for full scale operations at PFP. The interior volume of the laboratory furnace is approximately 1/10<sup>th</sup> the size of the production furnaces. The airflow in the laboratory furnaces for direct oxidation testing was scaled down in proportion to the interior volume. Calculations for full scale operation have been based on stabilizing two polycubes containing 400 grams polystyrene per furnace cycle.

Since the weight loss by the polycube is due to pyrolysis (approximately 50%) and oxidation, any calculations that ignores the contribution of the oxidation to the weight loss is conservative in the estimate of flammable gases concentrations. Another significant observation from the testing that will influence the flammability of the oxidation process is that as the weight of the sample increases, the relative rate of weight loss decreases. Thus, the flammable gases generation rate will be lower for full sized polycubes compared to what would be expected from a linear extrapolation based on rates measured for the small samples used in the TA MS experiments.

Surface Area Effect

The effect of surface area on flammable gas generation from polycubes during direct oxidation is very difficult to determine. When polystyrene is heated, it melts and the surface area changes. At the same time, the oxidation and pyrolysis processes are occurring. Tests on pure polystyrene in the PPSL have determined that as the weight of sample increases, the relative rate of weight loss decreases. When radiation of polystyrene causes the surface area to increase in the polycubes, oxidation is also occurring. The latter two processes have the counteracting effect of decreasing flammable organic gases generation and will nullify, if any, the effect of increased surface area. This will have the effect of counteracting the effect of increased surface area. The melting temperature of polystyrene is much lower than the temperature pyrolysis occurs at a significant rate. The melting of the damaged polycube will fuse the smaller particles, effectively reducing the surface area.

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Flammability Limit of Mixtures

The most reliable method of determining flammability limits for mixtures is to experimentally determine the value for specific mixtures. The organic compounds that are produced as a result of pyrolysis individually have lower flammability limits (LFLs) of approximately 1% by volume in air. Carbon monoxide is produced through incomplete oxidation of carbon. The LFL for CO is 12.5%. For similar organic compounds, Le Chatelier's rule may be applied to estimate the LFL for the mixture. The amount of each organic compound and its LFL are scaled to produce the LFL for the mixture. Because carbon monoxide is significantly different from aromatic hydrocarbons, it seems inappropriate to apply Le Chatelier's rule to estimate the LFL for the mixture. On the other hand, there are no mechanisms present that would produce a lower limit for the mixture than for the individual compound with the lowest LFL.

For the purpose of evaluating the fire safety of the direct oxidation process, calculations were performed using 400 grams of polystyrene. When considering whether the process could safely operate at 25% of the LFL, the LFL value of 0.8% for ethyl benzene was selected. Ethyl benzene is the compound with the lowest LFL of all the flammable gases observed in experiments at LANL, PPSL, and PNNL. Using Le Chatelier's rule, the LFL for the mixture would be higher than 0.8%, so using this value is a conservative approach.

Maximum Flammable Gas Generation Rate

The highest flammable gas generation rate has been observed in small samples of pure polystyrene with no cross linking. The weight loss rate of samples is related to gas generation rate. Under inert atmosphere conditions, all of this weight loss may be attributed to pyrolysis gases. In TA-MS experiments using polystyrene cross linked with 8% divinyl benzene, increasing the sample size from 20 mg to 100 mg decreased the maximum weight loss rate from 21% to 15% per minute. The same material was used in larger scale muffle furnace tests. Samples weighing 10, 20, and 40 grams were heated rapidly in a muffle furnace flushed with nitrogen. The maximum weight loss rate for these larger samples decreased from 4.8% to 4% per minute as the sample size increased.

Large scale tests with actual polycubes were performed in flowing air atmosphere. The maximum weight loss rate observed for 200 grams of polycube was 1% per minute. In the presence of air, the weight loss is due both to oxidation and pyrolysis.

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**Conclusions**

- 1 Factors affecting the concentration of flammable gases produced during the process can be controlled
  - a Process design will control the charge size to the furnace and therefore the amount of polystyrene that may be present in the system
  - b The rates of oxidation and pyrolysis are temperature dependent. The rate of each process has different temperature dependence. Process testing will provide a temperature profile that will maximize oxidation while providing a sufficient throughput for the process.
  - c The temperature ramp rate and dwell times at temperatures can be controlled using the existing muffle furnace controllers.
  - d Airflow through the furnace can be adjusted to provide sufficient dilution of flammable gases.
- 2 The proposed process does not ignite the polystyrene as was the case during PFP processes utilized in the 1960s. As a result, plugging of the off gas system will not be a problem. In laboratory tests, a filter from the production furnaces was placed in the off gas line and did not plug.
- 3 There have been some concerns that the flammable gases produced will cool and condense in the off gas system. Calculations based on the rate of air flow and flammable gas generation rate have shown that the vapor pressures of the flammable gases are sufficiently high that condensation will not occur.
- 4 Ability of the oxide material produced to meet storage criteria has been demonstrated. The carbon char is destroyed under the oxidizing conditions used in the direct oxidation process.
- 5 The amount of material handling required is significantly lower for direct oxidation compared to the pyrolysis followed by oxidation process. Destruction of the polystyrene matrix and oxidation of the resulting char material is performed in a single furnace without having to move the material to a separate furnace. Due to the high radiation dose operators will receive when handling this material, any reduction in handling is significant and will have a direct bearing on the achievable stabilization schedule.
- 6 Using direct oxidation can be treated as a feed shift for the muffle furnace operations based on completion of the Safety Basis documentation rather than an entirely new process. This will eliminate the need to install new equipment in a new glovebox, write

**FLUOR DANIEL HANFORD, INC****INTEROFFICE CORRESPONDENCE**

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procedures for the new process and train operators to the new equipment. In order to use the pyrolysis process, a testing program would have to be developed to determine operating parameters for actual polycubes.

- 7 Using existing furnaces provides the ability to more effectively utilize lag times for stabilization of polycubes while operating other processes.
- 8 Thurman Cooper in the PPSL has modeled several upset conditions for the process including a runaway temperature event in the furnace, loss of vacuum to the furnace, and loss of ventilation in the glovebox. These scenarios have the potential to produce flammable concentrations of organic gases. The maximum flammable gas generation rate used in these models was obtained from 40 gram sample of pure polystyrene. The models show that the glovebox containment will not be compromised under conditions of failure of any two of these controls while processing 400 grams polystyrene.
- 9 Removal of the existing polycube coating is not necessary. Aluminum paint and plastic tape was used to coat polycubes so that they could be handled in the Critical Mass Laboratory without contaminating the worker or equipment. Previous recovery processes removed these coatings prior to plutonium recovery. The quality of plutonium was a significant factor at the time. With time, these coatings have degraded so that it would not be possible to remove the coating. Because the purity to the plutonium is no longer a concern, no effort should be expended to separate the coating material from the polycubes.

**Recommendations**

Based on analyses performed in the PPSL and at PNNL, using a batch size of 400 grams in existing PFP muffle furnaces, polycubes can be safely stabilized in a one step, direct oxidation process. The material produced in this process will meet the required storage criteria.

Laboratory testing to provide input for the safety analysis for direct oxidation and to determine optimum process parameters should continue. Technical reports with the results of these tests should be written.

**Department of Energy**

Washington, DC 20585

**FEB 16 2000**RECEIVED  
00 FEB 18 PM 3: 18  
DNF SAFETY BOARD

The Honorable John T. Conway  
Chairman  
Defense Nuclear Facilities Safety Board  
625 Indiana Avenue, N.W.  
Suite 700  
Washington, D.C. 20004-2901

Dear Mr. Chairman:

This letter informs you of the completion of Commitment 113, "Document a Decision for Polycubes Stabilization Path Forward," included in the Implementation Plan (IP) for the Defense Nuclear Facilities Safety Board (DNFSB), Recommendation 94-1, Remediation of Nuclear Materials in the Defense Nuclear Facilities Complex, Rev. 2. The enclosed documentation, *White Paper Supporting Decision to Stabilize Polycubes using Direct Oxidation in Muffle Furnaces*, provides supporting preliminary technical details related to the above-mentioned decision.

The selected path forward for polycube stabilization at the Plutonium Finishing Plant is to use a one-step muffle furnace process instead of a two-step process, i.e., pyrolysis followed by muffle furnace stabilization. This path forward will provide significant opportunity for cost reduction and schedule recovery. Additional testing is required to develop processing parameters necessary to determine estimated throughputs within acceptable limits. After integration with the balance of plant activities, a schedule will be developed by February 29, 2000, to support the IP commitment to determine a completion date for polycube stabilization.

We continue to closely track progress on all Recommendation 94-1 commitments and will keep you and your staff apprised of our progress. If you have any questions, please contact me at (202) 586-5151 or Mark W. Frei, Deputy Assistant Secretary for Project Completion, at (202) 586-0370.

Sincerely,

A handwritten signature in black ink that reads "David G. Huizenga".

David G. Huizenga  
Deputy Assistant Secretary  
for Integration and Disposition  
Office of Environmental Management

Enclosure

cc w/enclosure:  
M. Whitaker, S-3.1



RL-F-1325.6 (02/98)

United States Government

Department of Energy  
Richland Operations Office

# memorandum

DATE: FEB 7 2000  
REPLY TO: MDD:MRH/00-MDD-036  
ATTN OF:

SUBJECT: PLUTONIUM FINISHING PLANT (PFP) COMPLETION OF DEFENSE NUCLEAR FACILITIES SAFETY BOARD 94-1 IMPLEMENTATION PLAN, REV. 2, COMMITMENT 113, "DOCUMENT A DECISION FOR POLYCUBES STABILIZATION PATH FORWARD"

TO: David G. Huizenga  
Deputy Assistant Secretary  
for Integration and Disposition,  
EM-20, HQ

- References:
- (1) FDH letter, A. Clark, J. L. Buel, L. J. Olguin to P. M. Knollmeyer, "White Paper Supporting Decision to Stabilize Polycubes Using Direct Oxidation in Muffle Furnaces" (9959367), dated December 29, 1999.
  - (2) RL memorandum, K. A. Klein to C. L. Huntoon, EM-1, "Proposed Revision to the Plutonium Finishing Plant Portion of the Defense Nuclear Facilities Safety Board Recommendation 94-1 Implementation Plan" (00-MDD-016), dated November 8, 1999.

This memorandum reports completion of the Implementation Plan (IP) for the Defense Nuclear Facilities Safety Board (DNFSB), Recommendation 94-1, Remediation of Nuclear Materials in the Defense Nuclear Facilities Complex, Rev. 2, Commitment 113, "Document a Decision for Polycubes Stabilization Path Forward," due date for the commitment was January 2000. Reference 2 provided a proposed revision to the PFP portion of the DNFSB Recommendation 94-1 IP which includes adding the subject commitment. Actual completion date for the commitment was December 29, 1999.

A one-step muffle furnace process will be employed to stabilize polycubes (Reference 1). This approach will provide significant opportunity for cost reduction and schedule recovery. Additional testing is required to develop processing parameters necessary to determine estimated throughputs. After integration with the balance of plant activities, a schedule will be developed to support the proposed IP Commitment to determine a completion date for polycube stabilization by February 2000.



David G. Huizenga  
00-MDD-036

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FEB 7 2000

The stabilization process utilizing the pyrolysis equipment developed by Los Alamos National Laboratories (LANL) will be retained as a backup alternative. The equipment has been crated up and may be shipped to Hanford for storage.

If you have any questions, please contact me, or your staff may contact Peter M. Knollmeyer, Assistant Manager for Nuclear Materials and Facility Stabilization, on (509) 376-7435.



Keith A. Klein  
Manager

cc: C. J. Sink, Jr., EM-21  
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**FLUOR DANIEL**

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December 29 1999

FH-9959367

Mr P M Knollmeyer Assistant Manager  
Nuclear Materials and Facility Stabilization  
U S Department of Energy  
Richland Operations Office  
P O Box 550  
Richland Washington 99352

Dear Mr Knollmeyer

**WHITE PAPER SUPPORTING DECISION TO STABILIZE POLYCUBES USING  
DIRECT OXIDATION IN MUFFLE FURNACES**

Reference Memo S A Jones FH to R K Leugemors FH same subject  
15F00-99 168 dated December 29 1999

Fluor Hanford has reviewed the option for processing the Plutonium Finishing Plant (PFP) inventory of Plutonium (Pu) polycubes by thermal stabilization (muffle furnace) without prior pyrolysis processing. We have determined that the current or similar muffle furnaces in use at PFP can and will be utilized to safely and more efficiently stabilize these items.

This decision is based on a technical evaluation performed by the Plutonium Plant Support Laboratories (PPSL) as documented in the above Reference which incorporates the results of testing and analysis completed to date by PPSL and the Pacific Northwest National Laboratories. This report documents that polycube processing can be performed at a 400 gram loading and still maintain the peak flammable off gas below 25 percent of the lower flammability limit. This loading combined with conservative process cycle periods indicates that thermal stabilization can be performed more efficiently than pyrolysis. This will also eliminate the expenditure for new equipment installation and will support an earlier startup of polycube stabilization.

A Kepner Trego evaluation was performed on December 22 1999 by a diverse group of technical and project personnel. The review included a comparison of direct thermal processing vs pyrolysis followed by thermal processing in the following areas: safety, ability to meet 3015 and Materials Disposition Standards, ALARA, technical confidence, schedule, cost, facility logistics, process reliability, and maintainability. The



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results showed that direct thermal stabilization was clearly preferential to pyrolysis followed by thermal stabilization especially in the categories of safety, schedule and cost

This letter closes the interim commitment to identify the polycube stabilization process as identified in the anticipated December 1999 update of the Defense Nuclear Facility Safety Board (DNFSB) 94-1 Implementation Plan. A revised schedule for completion of PFP polycube stabilization will be developed based on this selected path forward for polycube processing the results of subsequent polycube testing at PFP and PNNL and integration of the revised path forward with other DNFSB 94-1 commitments

If you have any questions or require additional information please contact Mr R K Leugemors at 373 0729

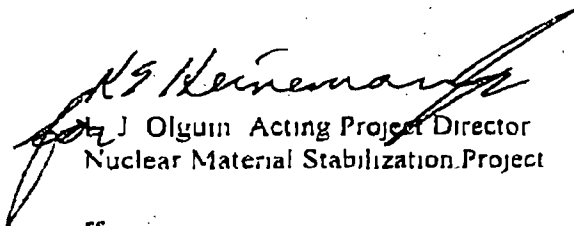
Sincerely



A. Clark Senior Director  
Project Planning and Integration  
Nuclear Material Stabilization Project



J. L. Buelt Product Line Manager  
Process and Measurement Technology  
Pacific Northwest National Laboratory



J. Olguin Acting Project Director  
Nuclear Material Stabilization Project

rs

Attachment

ATTACHMENT  
FH-9959367**FLUOR DANIEL HANFORD, INC****INTEROFFICE CORRESPONDENCE**

To R K Leugemors T2 12 Date 15F00 99 168  
December 29, 1999

From S A Jones *S Jones* T5-12 Telephone 373 3347

cc Distribution

Subject WHITE PAPER SUPPORTING DECISION TO STABILIZE POLYCUBES USING  
DIRECT OXIDATION IN MUFFLE FURNACES

- References (1) Lewis, W S, "Stabilization of Polycubes Engineering Study",  
WHC SD CP-TI-204, September 1996, Westinghouse Hanford, Company,  
Richland, WA
- (2) Jones, S A, G S Barney, and J Abrefah 'Muffle Furnace Stabilization of  
Polycubes', September 9, 1999 B&W Hanford Company Richland, WA

**Purpose**

As part of stabilizing the inventory of plutonium at the Plutonium Finishing Plant, polycubes must be treated to produce plutonium and uranium oxides in a form suitable for storage. This white paper will provide the technical justifications for the decision to stabilize polycubes using muffle furnace oxidation. Detailed technical reports will be issued separately for the laboratory studies and calculations that have been performed to support this decision.

**Background**Origin of Polycubes

Nuclear criticality studies at Hanford in the 1960's and 1970's used polycubes in order to prepare items with fixed H/X ratios. The ratio of hydrogen to fissile atoms affects neutron moderation. Using these solid forms, simulated "solutions" that were easy to handle and contained high concentrations of fissile material could be prepared. Polystyrene powder was mixed with plutonium and/or uranium oxides and heat pressed into "cubes". The dimensions were 2 inches wide by 2 inches deep with heights of 1/2, 1, 1 1/2, or 2 inches. The concentration of plutonium was such that H/X ratios of 5, 15, or 50 were achieved. Most of the polycubes remaining in the Hanford inventory had an H/X ratio of 50 before radiolytic decomposition.

Polystyrene Thermal Degradation

When polystyrene is heated in an inert environment, pyrolysis of polystyrene produces styrene monomer, toluene, ethyl benzene, and other alkyl benzene compounds. In the inert environment,

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all of the polystyrene is converted to hydrocarbons and a carbon char. Destruction of the char requires heating in an oxidizing atmosphere.

When heated in air, pyrolysis also occurs to produce styrene, toluene, and ethyl benzene but some of the polystyrene is oxidized producing water, carbon monoxide, and carbon dioxide. The thermal decomposition of polystyrene in air, generating these gaseous products is strongly influenced by temperature and the temperature ramp rate. A change of temperature not only causes the reaction rate to change but also changes the mechanism. Oxidation of polystyrene begins at a lower temperature than pyrolysis.

Historical Treatment

Up until about 1985, the plutonium in polycubes was recovered in the Plutonium Reclamation Facility (PRF). From the inventory of material at the Plutonium Finishing Plant, it appears the highest plutonium concentration cubes were processed. What remains today are approximately 1600 polycubes containing about 35 kg of plutonium. Most of these polycubes appear to be of similar composition. This conclusion was reached by examining the item identification numbers, the plutonium content of the individual items, and the age of the items.

The first method to recover plutonium from polycubes involved burning polycubes in the Plutonium Reclamation Facility. Any tape and paint coating was removed from the polycubes. Using a hammer and cold chisel the polycubes were broken up into smaller pieces and placed into a Hoskins type furnace at 600 °C where they burned. During heating, air was pulled through the furnace and four wires maintained at red heat above the crucible ignited vapors. Decarbonization was considered complete when there was no longer any flame or glowing in the crucible. This process produced plutonium oxide of acceptable quality. However, this process produced considerable amounts of soot that plugged off gas filters. The temperature was difficult to control once the ignition temperature of the styrene monomer was reached. Controlling the temperature was significant in this time, because the higher fired oxides were more difficult to dissolve. This plutonium recovery was being performed during a time when plutonium weapons production was still active and the quality of the oxide for this purpose was an important consideration.

A new process developed in 1969 consisted of removing the coatings, distilling styrene monomer in an inert environment, and decarbonizing the oxide product of the distillation. This method was discontinued due to many problems including inadequate collection of the styrene monomer in a condenser. During this operation, styrene monomer was observed outside the distillation apparatus condensing and running down the interior glovebox walls.



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#### Reason for Stabilizing Polycubes

The current storage configuration for polycubes is unacceptable for long term storage. Because radiolytic decomposition of polystyrene by the decay plutonium generated gases that pressurized sealed containers, these items are stored in a vented configuration. Radiolysis has degraded the polystyrene. When the polycubes were first placed in these storage containers they could be handled with little concern for contamination control because a coating of paint and tape sealed the polystyrene and plutonium cubes. Containers were vented and a filter placed over the opening so that gases produced by radiolysis could escape while maintaining contamination control. By 1985, the polystyrene had degraded enough that in handling the containers and inverting them, an air borne contamination was produced. Decomposition of the glue holding the filter in place allowed radioactive particles to escape from the storage container during inversion of the storage container. The filters glued over the opening are adequate for storage when the items are not moved.

#### Effect of Radiation on Polystyrene

Pure polystyrene and the polystyrene in actual polycubes differ because of the effect of radiation on the polystyrene. High energy radiation is nonspecific and all chemical bonds have a certain probability of involvement, which depends on their electron density. The process rarely consists only of chain scission, usually cross linking occurs simultaneously. One effect of radiation on polystyrene is to cause it to lose hydrogen. The initial effect is to produce cross linking. Cross linking of the polystyrene in polycubes has lowered their mechanical strength and has caused the polystyrene to have a higher pyrolysis temperature.

Radiation damage to polystyrene increases radical production, which increases the rate of polystyrene oxidation. Polymer oxidation reactions generally show a slow induction period during which no visible changes occur in the material. Oxidation leads to decreased molecular weight and discoloration of the polymer. In the presence of air, auto oxidation also occurs through reactions with atmospheric oxygen. These polycubes were stored in vented containers to prevent pressurization from the release of hydrogen. This means that oxygen was present. Evidence of oxidation is seen in that the polycubes have become friable.

A report of results of critical experiments performed with polycubes having an H/Pu ratio of 50 listed the isotopic abundances of the plutonium isotopes that had a  $^{240}\text{Pu}$  isotopic content of 18.35% in 1970. Calculating the decay of  $^{241}\text{Pu}$  and the in growth of  $^{241}\text{Am}$ , these polycubes now contain approximately 5%  $^{241}\text{Am}$  relative to total plutonium. This americium concentration is consistent with the high radiation doses reported on the containers of polycubes.

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Calculations based on the amount of plutonium in the polycubes in contact with polystyrene for 30 years yield that the polycubes would have been exposed to a dose of 10 to 100 megarads during their storage. This dose is sufficient to have reduced the hydrogen content in the polycubes significantly. By comparison, freshly prepared polyethylene polycubes with an H/X ratio of 15 decreased to an H/X ratio of 14.28 in 98 days. Based on the 30 years since the preparation of these polycubes, the amount of hydrogen in the system is significantly lower than was originally present.

As a result of radiolysis, the amount of hydrogen containing carbon compounds is lower in irradiated polycubes compared to pure polystyrene. Consequently, the flammable organic compounds generated in thermal stabilization of degraded polycubes will be lower compared to that observed in pure polystyrene.

Historical Baseline Using Pyrolysis

The current baseline process was selected based on an engineering study of polycube stabilization that concluded the pyrolysis method developed at Los Alamos National Laboratory (LANL) with a silent gas plasma off gas treatment was the preferred alternative (Lewis, 1996). The off gas treatment was later changed to a catalytic oxidation process. The pyrolysis method involves a device that heats the polycubes to 750 °C in an inert atmosphere. Flammable organic products of the pyrolysis are collected in a reservoir. After accumulating sufficient quantity of these flammable compounds, a series of pumps and valves feed the compounds and air into a catalytic converter that oxidizes the hydrocarbons. Monitors for hydrocarbons provide controls for the process through a computer feedback system. Although considerable effort has been invested at LANL into the design and optimization of this process using pure polystyrene, no tests were conducted using actual polycubes. The residue from the pyrolysis step must be transferred to a muffle furnace where the carbon char is oxidized to complete the stabilization process. This stabilization process was identified as the baseline in the 1999 submittal of the PFP Integrated Project Management Plan (IPMP).

Proposed Baseline Change to Direct Oxidation

Studies of the gases generated when polystyrene was heated in air were performed in the PFP Plutonium Process Support Laboratories (PPSL) as part of the plutonium sludge stabilization safety analysis. It was found that when polystyrene was heated in flowing air, a significant portion of the polystyrene was directly oxidized without combustion. Similar results were identified during initial tests at PNNL conducted for evaluation of the LANL pyrolysis processes. The results of these studies indicated that it might be possible to safely stabilize polycubes using the existing muffle furnaces at PFP. Additional studies were performed in the PPSL and at Pacific Northwest National Laboratory (PNNL). The results of these studies are the

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basis of the decision that muffle furnace oxidation should be used rather than pyrolysis for the stabilization of polycubes

Description of Current Muffle Furnace Operations

The muffle furnaces currently used for stabilization of plutonium oxides scraps, and sludges have an internal furnace volume of approximately six liters. A programmable temperature controller is used to adjust the temperature ramp rates including dwell times at fixed temperatures. Air is drawn in through the space around the loosely fitted door of the furnace using a vacuum source at the back of the furnace separate from the glovebox ventilation. For some feed material carbon dioxide is added to the air in the furnace to reduce the potential for a flammable gas mixture developing in the furnace. Feed material is placed in stainless steel boats and then inserted into the furnace. After the heating cycle is completed the furnace is allowed to cool before the boat is removed.

**Direct Oxidation Process**

An accumulation of organic vapors above the lower flammability limit inside a plutonium handling glovebox is an unacceptable situation. LANL pyrolysis equipment uses an argon atmosphere to exclude oxygen during the pyrolysis. The hydrocarbons generated by that process are oxidized in a separate step using a metering pump and valves to control the amount of hydrocarbons and air combined at any time.

The proposed process for direct oxidation of polycubes in muffle furnaces will produce flammable gases in the air at very low concentrations. Tests were designed to measure the rate of flammable gas generation and the composition of the gases resulting from direct oxidation of pure polystyrene, simulants and actual polycube samples.

Testing at Hanford was conducted in three areas:

1. At PPSL, G Scott Barney tested synthetic polycubes using polystyrene, polystyrene mixed with cerium oxide to simulate the effect of plutonium oxide, and polystyrene cross linked with divinyl benzene to simulate the effect of radiation on polycubes. These tests were performed on milligram scale tests in a Thermal Analyzer Mass Spectrometer (TA MS) system and on multigram samples in a muffle furnace.
2. At PNNL, a very similar TA MS system was used by John Abrefah to test milligram-sized samples of actual polycubes. These actual polycubes were selected as ones that were representative of the inventory of polycubes to be stabilized.
3. Also at PPSL, Susan Jones tested full sized, actual polycubes in a muffle furnace. These polycubes contained less radioactivity than the ones tested at PNNL. These are thought to be the worst case polycubes.



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A number of conservative assumptions have been made for the safety analysis. Pure polystyrene was used for calculation of the maximum gas generation rate. Experiments using varying degrees of cross linking in pure polystyrene have demonstrated that the gas generation rate decreases with increasing cross linking. Additionally, experiments with the actual polycubes at PNNL and PPSL show that the rate of weight loss is slower than with pure polystyrene. This has been demonstrated using small (up to 600 mg) samples in the TA-MS and using full sized polycubes (up to 218 g) in the laboratory muffle furnace.

The laboratory muffle furnaces are much smaller than the units used for full scale operations at PFP. The interior volume of the laboratory furnace is approximately 1/10<sup>th</sup> the size of the production furnaces. The airflow in the laboratory furnaces for direct oxidation testing was scaled down in proportion to the interior volume. Calculations for full scale operation have been based on stabilizing two polycubes containing 400 grams polystyrene per furnace cycle.

Since the weight loss by the polycube is due to pyrolysis (approximately 50%) and oxidation, any calculations that ignores the contribution of the oxidation to the weight loss is conservative in the estimate of flammable gases concentrations. Another significant observation from the testing that will influence the flammability of the oxidation process is that as the weight of the sample increases, the relative rate of weight loss decreases. Thus, the flammable gases generation rate will be lower for full sized polycubes compared to what would be expected from a linear extrapolation based on rates measured for the small samples used in the TA MS experiments.

#### Surface Area Effect

The effect of surface area on flammable gas generation from polycubes during direct oxidation is very difficult to determine. When polystyrene is heated, it melts and the surface area changes. At the same time, the oxidation and pyrolysis processes are occurring. Tests on pure polystyrene in the PPSL have determined that as the weight of sample increases, the relative rate of weight loss decreases. When radiation of polystyrene causes the surface area to increase in the polycubes, oxidation is also occurring. The latter two processes have the counteracting effect of decreasing flammable organic gases generation and will nullify, if any, the effect of increased surface area. This will have the effect of counteracting the effect of increased surface area. The melting temperature of polystyrene is much lower than the temperature pyrolysis occurs at a significant rate. The melting of the damaged polycube will fuse the smaller particles, effectively reducing the surface area.

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Flammability Limit of Mixtures

The most reliable method of determining flammability limits for mixtures is to experimentally determine the value for specific mixtures. The organic compounds that are produced as a result of pyrolysis individually have lower flammability limits (LFLs) of approximately 1% by volume in air. Carbon monoxide is produced through incomplete oxidation of carbon. The LFL for CO is 12.5%. For similar organic compounds, Le Chatelier's rule may be applied to estimate the LFL for the mixture. The amount of each organic compound and its LFL are scaled to produce the LFL for the mixture. Because carbon monoxide is significantly different from aromatic hydrocarbons, it seems inappropriate to apply Le Chatelier's rule to estimate the LFL for the mixture. On the other hand, there are no mechanisms present that would produce a lower limit for the mixture than for the individual compound with the lowest LFL.

For the purpose of evaluating the fire safety of the direct oxidation process, calculations were performed using 400 grams of polystyrene. When considering whether the process could safely operate at 25% of the LFL, the LFL value of 0.8% for ethyl benzene was selected. Ethyl benzene is the compound with the lowest LFL of all the flammable gases observed in experiments at LANL, PPSL, and PNNL. Using Le Chatelier's rule, the LFL for the mixture would be higher than 0.8%, so using this value is a conservative approach.

Maximum Flammable Gas Generation Rate

The highest flammable gas generation rate has been observed in small samples of pure polystyrene with no cross linking. The weight loss rate of samples is related to gas generation rate. Under inert atmosphere conditions, all of this weight loss may be attributed to pyrolysis gases. In TA MS experiments using polystyrene cross linked with 8% divinyl benzene, increasing the sample size from 20 mg to 100 mg decreased the maximum weight loss rate from 21% to 15% per minute. The same material was used in larger scale muffle furnace tests. Samples weighing 10, 20, and 40 grams were heated rapidly in a muffle furnace flushed with nitrogen. The maximum weight loss rate for these larger samples decreased from 4.8% to 4% per minute as the sample size increased.

Large scale tests with actual polycubes were performed in flowing air atmosphere. The maximum weight loss rate observed for 200 grams of polycube was 1% per minute. In the presence of air, the weight loss is due both to oxidation and pyrolysis.

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**Conclusions**

- 1 Factors affecting the concentration of flammable gases produced during the process can be controlled
  - a Process design will control the charge size to the furnace and therefore the amount of polystyrene that may be present in the system
  - b The rates of oxidation and pyrolysis are temperature dependent. The rate of each process has different temperature dependence. Process testing will provide a temperature profile that will maximize oxidation while providing a sufficient throughput for the process.
  - c The temperature ramp rate and dwell times at temperatures can be controlled using the existing muffle furnace controllers.
  - d Airtlow through the furnace can be adjusted to provide sufficient dilution of flammable gases.
- 2 The proposed process does not ignite the polystyrene as was the case during PFP processes utilized in the 1960 s. As a result, plugging of the off gas system will not be a problem. In laboratory tests, a filter from the production furnaces was placed in the off gas line and did not plug.
- 3 There have been some concerns that the flammable gases produced will cool and condense in the off gas system. Calculations based on the rate of air flow and flammable gas generation rate have shown that the vapor pressures of the flammable gases are sufficiently high that condensation will not occur.
- 4 Ability of the oxide material produced to meet storage criteria has been demonstrated. The carbon char is destroyed under the oxidizing conditions used in the direct oxidation process.
- 5 The amount of material handling required is significantly lower for direct oxidation compared to the pyrolysis followed by oxidation process. Destruction of the polystyrene matrix and oxidation of the resulting char material is performed in a single furnace without having to move the material to a separate furnace. Due to the high radiation dose operators will receive when handling this material, any reduction in handling is significant and will have a direct bearing on the achievable stabilization schedule.
- 6 Using direct oxidation can be treated as a feed shift for the muffle furnace operations based on completion of the Safety Basis documentation rather than an entirely new process. This will eliminate the need to install new equipment in a new glovebox, write

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procedures for the new process and train operators to the new equipment. In order to use the pyrolysis process, a testing program would have to be developed to determine operating parameters for actual polycubes.

- 7 Using existing furnaces provides the ability to more effectively utilize lag times for stabilization of polycubes while operating other processes.
- 8 Thurman Cooper in the PPSL has modeled several upset conditions for the process including a runaway temperature event in the furnace, loss of vacuum to the furnace, and loss of ventilation in the glovebox. These scenarios have the potential to produce flammable concentrations of organic gases. The maximum flammable gas generation rate used in these models was obtained from 40 gram sample of pure polystyrene. The models show that the glovebox containment will not be compromised under conditions of failure of any two of these controls while processing 400 grams polystyrene.
- 9 Removal of the existing polycube coating is not necessary. Aluminum paint and plastic tape was used to coat polycubes so that they could be handled in the Critical Mass Laboratory without contaminating the worker or equipment. Previous recovery processes removed these coatings prior to plutonium recovery. The quality of plutonium was a significant factor at the time. With time, these coatings have degraded so that it would not be possible to remove the coating. Because the purity to the plutonium is no longer a concern, no effort should be expended to separate the coating material from the polycubes.

**Recommendations**

Based on analyses performed in the PPSL and at PNNL, using a batch size of 400 grams in existing PFP muffle furnaces, polycubes can be safely stabilized in a one step, direct oxidation process. The material produced in this process will meet the required storage criteria.

Laboratory testing to provide input for the safety analysis for direct oxidation and to determine optimum process parameters should continue. Technical reports with the results of these tests should be written.