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Cost-Benefit Analysis of Unfired PuO_2 Pellets as an Alternative Plutonium Shipping Form

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ABSTRACT

A limited cost-benefit evaluation was performed concerning use of unfired plutonium dioxide pellets as a shipping form. Two specific processing operations are required for this use, one to form the pellet (pelletizing) and a second to reconstitute an acceptable powder upon receipt (reconstitution). The direct costs for the pelletizing operation are approximately $208,000 for equipment and its installation and $122 per kg of plutonium processed (based upon a 20-kg plutonium/day facility). The direct costs for reconstitution are approximately $90,000 for equipment and its installation and $81 per kg of plutonium processed. The indirect cost considered was personnel exposure from these operations. Whole body exposures ranged from 0.04 man-rem per 100 kg of low-exposure plutonium reconstituted to 0.9 man-rem per 100 kg of average-exposure plutonium pelletized. Hand exposures were much higher--17 man-rem per 100 kg of low-exposure plutonium reconstituted to 67 man-rem per 100 kg of average plutonium pelletized. The principal benefit is a potential twentyfold reduction of airborne release in the event of an accident. An experimental plan is outlined to fill the data gaps uncovered during this study in the areas of pelletizing and reconstitution process parameters and pellet response behavior to accident-generated stresses. A study to enhance the containment potential of the inner packaging used during shipment is also outlined.

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INTRODUCTION

BACKGROUND

Based upon one of the conclusions in its re-evaluation of present regulations covering air and other modes of transporting radioactive materials (NRC 1977), the U.S. Nuclear Regulatory Commission, Transportation and Materials Risk Branch, has contracted with the Pacific Northwest Laboratory (PNL) to study the feasibility of altering the current plutonium shipping form (PuO_2 powder) to reduce its dispersibility under shipping accident conditions. The initial phase of the study surveyed the current shipping practices and procedures, defined an envelope of accident conditions, and evaluated the feasibility of some candidate techniques and materials: green (unfired) pellets, sol-gel microspheres, inorganic ion exchangers, ablative materials and alternative packaging materials. A report on this phase (Mishima and Lindsey 1983) concluded that use of unfired PuO_2 pellets is the most promising approach. A recommendation of that report was to perform a limited cost-benefit analysis of this approach.

OBJECTIVES

The purpose of this report is to evaluate the potential costs and benefits of altering current shipping practice to use unfired plutonium dioxide (PuO_2) pellets in place of the PuO_2 powder now used. Two categories of potential costs will be evaluated:

direct costs such as capital (equipment), labor, redesign of process and transport procedures, resource commitment (supplies and services), additional transport costs

indirect costs such as additional personnel exposure during processing and transport.

Inasmuch as one of the selection criteria was to minimize changes in processing and shipping procedures, some of these costs are not applicable. The criticality implications of shipping a denser form of plutonium solid need to be addressed. Also, potential-injury statistics for additional processing (light manufacturing) were not considered.

The primary benefits considered are reduced dispersibility and potential downwind inhalation hazard of the PuO_2 under conditions defined in the accident envelope.

Since the techniques chosen have not been used for the specific purposes outlined here, questions arise as to how they can be best used to achieve the program objectives. An experimental program is required to obtain the values for various parameters needed to produce unfired pellets which exhibit the optimum normal and accident-generated stress response behavior and can be reconstituted into a satisfactory starting material for further use. Additionally, some techniques (outside the scope of this study) to further reduce the

potential dispersibility of unfired pellets, in the general area of inner packaging modification, were conceived during the study. In order not to lose any potential benefit resulting from the program, the information is being made available in this report.

SUMMARY AND CONCLUSIONS

A limited cost-benefit evaluation has been performed for the use of unfired plutonium dioxide pellets as an alternative plutonium shipping form. Unfired pellets were the most promising of the candidate techniques and materials considered in the feasibility study of alternative shipping forms that was the initial phase of this study (Mishima and Lindsey 1983). Direct costs considered in the current study were equipment, manpower, supplies and services for a throughput facility of 20 kg of plutonium per day for pelletizing and reconstituting the powder. The indirect cost considered was additional personnel exposure. Impacts of potential accidents and injuries were not covered. Increased potential for criticality from a denser plutonium form was considered but is considered irrelevant. Benefits are the reduced dispersion and inhalation hazard potentials in the unlikely event of a major accident during shipment.

The direct costs for pelletizing were determined to be approximately $208,000 (total) for equipment and installation and $122 per kg of plutonium for operations. The direct costs for reconstitution are approximately $90,000 for equipment and installation and $81 per kg of plutonium for operations. The additional personnel exposures for the two operations were calculated for use of average- or low-exposure plutonium: pelletizing (average) 67 man-rem to the hands and 0.9 man-rem whole body per 100 kg of plutonium, (low) 18 man-rem to the hands and 0.14 man-rem whole body per 100 kg of plutonium; reconstitution (average) 64 man-rem to the hands and 0.19 man-rem whole body per 100 kg of plutonium, (low) 17 man-rem to the hands and 0.04 man-rem whole body per 100 kg of plutonium. The major benefit is a twentyfold reduction of the potential airborne respirable fraction in the event of an accident, with a significant, but unspecified, reduction in dispersibility.

In the course of reviewing the literature for data to evaluate the feasibility of using unfired pellets as a plutonium shipping form and for this cost-benefit evaluation, it become apparent that the information available was not fully adequate for the task. Although the techniques to pelletize PuO_2 powders and to reconstitute the powder from the pellets are well established, both are used as interim process steps, and the measurements to define the characteristics of the interim products have not been made or documented. An experimental program is outlined with the objective of providing that data in the area of pelletizing and reconstitution process parameters to produce the strongest possible pellet that can be reconstituted into a satisfactory starting material for fuel fabrication. Such pellets would produce coarse fragments if subjected to accident-generated forces. An optional study of methods to enhance the containment capability of the inner packaging is also outlined.

DISCUSSION

In order to provide the information required for a limited cost-benefit evaluation, PNL evaluated the major elements influencing the cost and benefit. Three principal areas covered were direct costs, indirect costs, and benefits. Major elements within these areas considered were the direct cost of installing the techniques (pelletizing and reconstitution) and producing the products (unfired pellets and powder), the indirect cost (assumed to be personnel exposure during processing and handling), and the benefit (reduced potential dispersion and downwind inhalation hazard from transport of plutonium). Another possible direct/indirect cost, the potential criticality impact from the use of a denser plutonium form, was considered but was found to be irrelevant. Other potential costs (e.g., such as redesign of transport containers, additional facilities) were eliminated by the alternative form chosen. The technique to produce this form is currently in use; it requires only one additional process step (pelletizing) and uses the current powder shipped as a starting material. The techniques used for reconstitution of the starting material are currently in use at the type of facility (fuel fabrication plants) that would require them if the alternative form were adopted. The cost of installation was evaluated for the as-yet-undefined type of facility that may need such capability in the future. Air cleaning system costs were not included since it was assumed that any plant handling plutonium would already have this capability. Other indirect costs were not evaluated (e.g., potential injuries due to additional processing required, potential additional risk from accident consequences, etc.) because they are believed to be small and the information is readily available.

Although the techniques outlined are available in the current technology, they have not been evaluated for the specific uses discussed in this study--the production of unfired pellets which generate the smallest fraction of fine particles under normal and accident conditions during transport and can be reconstituted into a satisfactory starting material for the intended use. An experimental plan to obtain such process parameters and pellet response behavior information is discussed, but its costs were not evaluated. It is recognized that these costs may well be the most significant costs in implementing use of this alternative form. The demands of such an evaluation (a detailed matrix of all the experiments is required to evaluate the man-hours, equipment costs including modifications for use with plutonium, and ancillary services required, such as chemical analysis and radiation monitoring) are beyond the scope of this limited study. A further cost, requalification of the fuel pellets, is eliminated by making it one of the objectives of the experimental plan.

Finally, some ideas in the area of inner packaging modification which could also aid in reducing the potential dispersibility of plutonium transported as unfired pellets suggested themselves during consideration of various aspects of this study. They are also presented.

COSTS

There are both direct and indirect costs associated with conversion from shipment of plutonium dioxide powder to that of unfired pellets. The direct costs are associated with process steps required to form the pellets for shipment and to reconstitute the powder upon its receipt. In the cases where the plutonium is considered a waste form, the comminution step will probably not be required, and other processes will be used to adjust the form for proper disposition. The process steps outlined and evaluated would be in addition to the process currently envisioned for segregation of the plutonium. The techniques required have been developed and are currently in use, so equipment design is not required. Development of various process parameters to produce the desired pellet and powder characteristics will be needed, and those identified are discussed in the next section. The cost estimates are based on a daily processing of 20 kg of PuO_2. Consolidation of the plutonium dioxide powder allows the shipment of 1.75 kg per one sheet metal can 3 1/2 in. in diameter and height. Redesign or modification of existing shipping containers is not required, nor are additional shipments necessary. The effect on shipping criticality concerns due to the use of higher-density materials is briefly addressed.

The indirect cost addressed is the additional personnel exposure during pelletization and reconstitution. Additional shipments are not required, and no additional exposure is postulated. Estimates of injury statistics were thought to be small and so were not considered. Potential severe accident risks were beyond the scope of the study, but such information is available in EISs and SARs for mixed-oxide fuel fabrication plants.

Direct Costs: PuO_2 Powder Pelletizing and Reconstitution

The objectives of this section are to describe the process operations for both the fabrication of the unfired plutonium dioxide pellets and reconstitution of powder from the pellets. It also presents cost estimates, both capital and operational, for these additional process operations.

Since the process for fabrication of the unfired pellets was specified as capable of pelletizing PuO_2 produced by "representative" fuel reprocessing facilities, a section has been included on the synthesis and characteristics of plutonium dioxide powder. After separation, the plutonium is in solution as nitrate and is transported in this form to the front end of the oxide production line.

Powder Synthesis and Characteristics

If sizable quantities of plutonium are scheduled for conversion, the nitrate solution is blended in 10-kg increments to provide uniform nitrate feed for conversion to plutonium oxide lots of approximately 100 kg. Sampling of the oxide for quality control and certification is done on each 10-kg batch produced.

The conversion to oxide can be accomplished by several different processes; and, as one would expect, the powders produced and the pellets pressed from the powders will have differing characteristics and properties, depending upon the conversion process used (Houston 1964). However, the properties of powders produced by a single process also can vary significantly, depending upon the processing parameters.

The oxalate process probably has been used most extensively for the production of plutonium dioxide. The first operation in this process is the precipitation of plutonium oxalate by combining the plutonium nitrate solution with oxalic acid. The precipitated plutonium oxalate crystals are separated from the liquid by filtration and heated in air to decompose the oxalate to the oxide which, if for ceramic use, is heated to a higher temperature to lower the surface area for ease of processing.

The precipitated plutonium oxalate crystals are the precursors of the plutonium oxide powder particles. They will determine the oxide particle size and morphology because these oxalate crystal characteristics are essentially unaffected during decomposition. Therefore, precipitation parameters such as temperature, solution concentrations, precipitation rate, agitation rate, and digestion time, which can affect the size and morphology of the oxalate crystals, will likewise affect these same properties of the derived oxide powder particles.

The compressibility of the oxide powder is affected more by the calcination temperature than by particle size and morphology because this temperature determines surface area and degree of particle agglomeration of the powder. Powders with extremely high surface areas can be impossible to pelletize, even with the addition of binder and lubricant. With some powders, high surface area accompanies extremely small particles (submicron) with highly irregular, textured surfaces. However, oxalate-derived oxide powder with surface areas above 50 m^2/g can be composed of particles in the 1 to 5 μm range. These particles could not have such large surface areas if only the external surfaces were contributing. Surface areas of this magnitude (50 m^2/g) for particles of this relatively large size are possible because oxalate decomposed at low temperatures produces a porous oxide particle; thus, a large portion of the powder surface measured by gas absorption is interior porosity. Due to this internal porosity, it is possible to pelletize powders with surface areas higher than would be feasible for oxides derived from other precursors.

Because of the possible variations in plutonium oxide powder sources and characteristics, the process operations included in this report for the fabrication of green pellets were selected with the objective of accommodating the widest range of powder characteristics. Some powders could be of a quality such that granulation and addition of binder/lubricant would not be required. On the other hand, it is possible that a powder could be encountered that would not be amenable to pelletizing by this process without some additional processing operation on the powder, such as additional calcining to reduce surface area or blending binder with the powder before slugging.

The use of organic binder and lubricant in the fabrication of plutonium oxide pellets may result in some degradation of the unfired pellets if they are stored for long periods before powder reconstitution.[a] The organics may undergo some decomposition from radiolysis by the plutonium radiation, resulting in weakened pellets. Therefore, it would be advantageous to omit the organics if pelletizing can be accomplished without them. Otherwise, the amount of organic used should be the minimum required to pelletize the powder and, if used, pellet degradation as a result of the radiolysis must be a consideration in the allowable storage time before or after the pellets are shipped.

An operator experienced in the fabrication of pellets will be able to press two or three slugs from a powder and judge whether the powder will require granulation and lubrication to form pellets of good integrity. In this way, the fabrication procedure can be modified to fit the powder being processed. It is assumed that the pellet produced must be free of laminations and cracks and be strong enough to handle without damage.

Pelletizing Process

The process operations specified for this cost estimate, for both the pelletizing and the powder reconstitution, were kept as simple as possible to minimize worker exposure from material handling. A simplified schematic diagram for the pelletizing process (Procedure 1) is shown in Figure 1. The

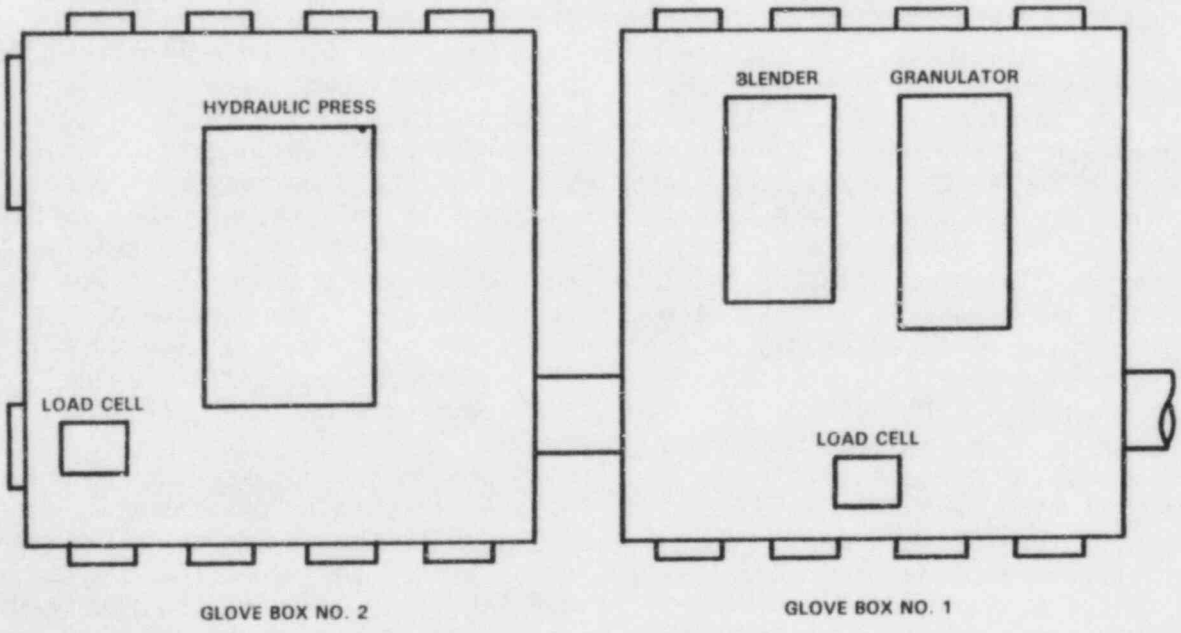

FIGURE 1. Glove Boxes for Fabrication of Unfired Plutonium Dioxide Pellets

(a) Paul Densley, Westinghouse Hanford Company, Richland, Washington. Personal Communication.

size of the pellet (1.5-in. dia) is large, so as to reduce the number of pellets per unit of powder as much as is practicable. The large pellet size reduces the number of shipping containers required for a given quantity of plutonium oxide and also limits the handling operations of the pelletized oxide. In addition, because the large pellet diameter is specified for the shipping form, the press tooling is also ideal for the slugging operation.

PROCEDURE 1. Process Operations for the Fabrication of Plutonium Dioxide Green Pellets.

1. Receive powder: ~ 1 kg in cans 3.5 in. dia x 3.5 in. high.

2. Confirm can weight.

3. Press slugs: Size - 1.5 in. dia x 0.5 to 0.4 in. high
   nominal 45.6 g/slug @ 41% theorotical density.

4. Granulate slugs: -14 mesh.

5. Blend granules with 0.25% stearic acid lubricant.

6. Press pellets from lubricated granules:
   Pellet size - 1.5 in. dia x nominal 0.87 in. high
   Nominal 144 g/pellet @ 50% TD.

7. Insert pellets into stainless-steel, closed-end cylinders of 1.51-in. dia: 4 pellets/cylinder.

8. Weigh each cylinder for net plutonium dioxide weight.

9. Package three cylinders per slip-lid can 3.5" dia x 3.5" high: Nominal 1.73 kg plutonium dioxide per can.

10. Obtain gross weight of can and net weight of plutonium dioxide.

The floor plan of the glove boxes in Figure 1 shows placement of the required equipment. The cans of powder with a maximum total plutonium oxide weight of 5 kg are received in Box No. 1. All metal shipping containers, both the cans and pellet cylinders, must be tared before introduction into the glove box. Upon receipt of the powder (normally, 5 cans for containing 5 kg of plutonium dioxide), each can is placed in a one-can rack in a corner of the box except the final can, which is weighed and transferred to the feeder shoe hopper on the press in Box No. 2. After slugging, the material is transferred (contained in the original can) to the granulator and granulated in Box No. 1. The second can of powder, weighed during the slugging of the first, can then be transferred to the press and slugged.

Meanwhile, the granulated slugs are returned to their original can, weighed, and stored in one of the one-can racks. In this way, only one can is

in process in a glove box at a time, but two operations are proceeding simultaneously until all five cans of powder have been slugged and granulated. The required amount of organic lubricant is then added to the granules, and the combined materials are blended in the can for ten minutes. Each can of granule/lubricant or press feed is transferred to Box No. 2 and pressed into pellets. The green pellets are inserted into the cylinders; the cylinders are weighed and then placed in the slip-lid cans, three cylinders per can. After a final weighing of the loaded can, it is stored in a one-can rack in Box No. 2 until all 5 kg of oxide have been fabricated into pellets and loaded into cans. The 5 kg of powder will produce three pellet-loaded cans for shipment. Bag-out can proceed after all 5 kg of pellets are loaded.

Estimated production rate for these process operations, employing two operators per shift, is 20 kg per day when operations are conducted on a basis of three eight-hour shifts per day. This mode of operation would allow approximately four hours during the day for glove box and equipment maintenance and cleanup.

The maximum design production rate of plutonium dioxide for the PUREX process at Hanford is 20 kg per day.[a] Plutonium production rates that were projected for West Valley[b] and Barnwell (Thomas 1979) were less, approximately 5 kg plutonium as nitrate and 12 to 14 kg of plutonium as mixed oxide, respectively. Therefore, it is believed that the unfired pellet fabrication capacity of 20 kg per day specified here is adequate and realistic.

Powder Reconstitution

Process operations for reconstitution of the powder are listed in Procedure 2, and a floor plan of the glove boxes with equipment placement is shown in Figure 2. These operations will require only one operator for four hours to process 5 kg of pellets.

PROCEDURE 2. Process Operations for the Reconstitution of Plutonium Dioxide Powder

1. Receive green plutonium dioxide pellets: nominal 1.73 kg per can.
2. Confirm can weight.
3. Granulate pellets.
4. Air-calcine granules for organic removal: 5 kg/run.

(a) Rick Hoyt. PUREX Project, Rockwell Hanford Operations, Richland, Washington. Personal Communication.
(b) W. R. Jacoby. West Valley Decontamination and Decommissioning Project, West Valley, New York. Personal Communication.

5. Ball mill granules: 2.5 kg/mill.

6. Unload mills.

7. Can powder: slip lid cans, 3.5 in. dia x 3.5 in. high; maximum 2.0 kg/can

8. Weigh to determine net weight of powder.

The three cans of pellets are weighed upon receipt into Box No. 1 for weight confirmation and stored in one-can racks at the corners of the box. The pellets from each can are granulated and the granules placed in a ceramic tray. The tray is then transferred to Box No. 2 and loaded into the furnace.

When these operations are completed for each can of pellets, the furnace is heated to 500°C and held at this temperature for four hours with air flowing over the granules to completely remove organics. The furnace is allowed to cool, and the trays are transferred back to Box No. 1 individually. The granules from the three trays are divided equally between two rubber-lined mills containing 0.5 in. stainless-steel balls. If the mills contain the proper volume of balls (between 0.4 and 0.5 of the mill volume) and are rotated at the correct rate for the diameter of the mill, the granules should be reduced to the original powder particle size in four hours. After milling, the

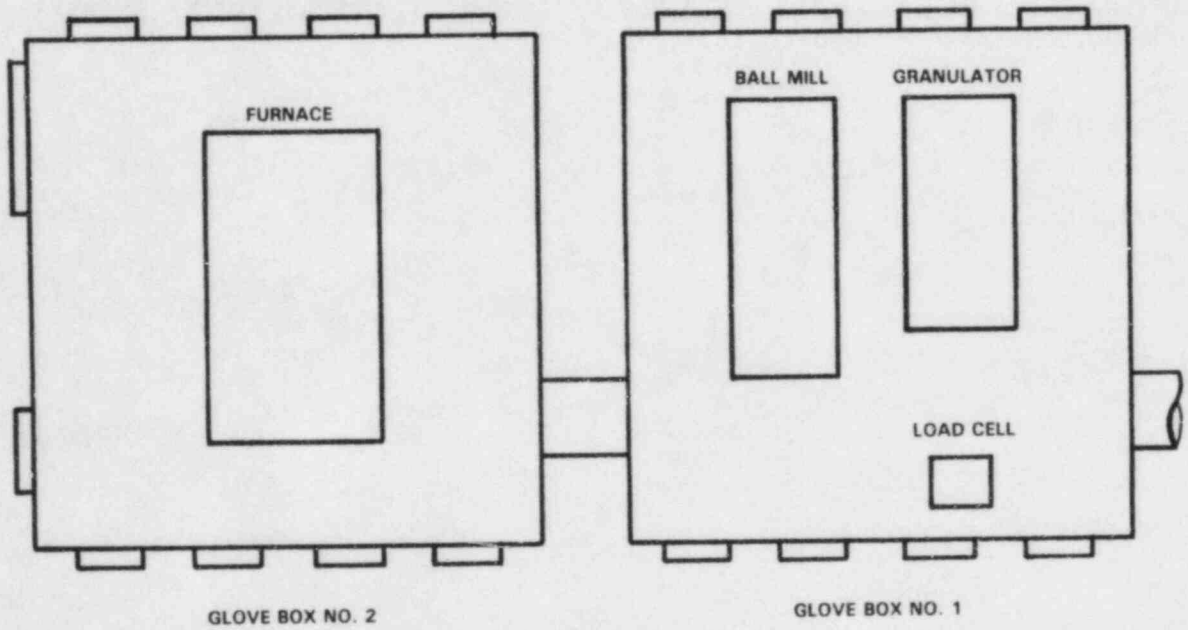

FIGURE 2. Glove Boxes for Reconstituting Unfired Pellets to Plutonium Dioxide Powder

powder should be reconstituted with the original particle size and approximately the same surface area, unless the initial powder was calcined at a temperature below 500°C. However, it is unlikely that the bulk and tap densities of the reconstituted powder will be the same as the original.

If the reconstituted powder will require special properties for subsequent use, such as high reactivity for hydrofluorination, the pelletizing and powder reconstitution processes may need modification to accommodate these special requirements. These modifications can be determined only by experimental development. It would be impossible to predict with certainty the properties of the reconstituted powder compared to the original.

The furnace cycle for organic removal from 5 kg of oxide granules will be a maximum of twelve hours; therefore, it is estimated that four continuous eight-hour shifts will be required to reconstitute (in ball mill and furnace) 10 kg of powder since only 5 kg of oxide are in process at one time.

Cost Estimates

The capital costs (Tables 1-5) were obtained by contacting suppliers for the items of equipment required for the various operations. Glove boxes were priced by the manufacturer and also by discussing glove box costs with a manager[a] at the Los Alamos Plutonium Laboratory. Glove box installation costs were estimated based on costs of installing similar boxes at the Los Alamos Plutonium Laboratory. However, labor cost estimates are based on the April 1983 rates at PNL. All equipment prices are for March to May 1983 and include purchasing-overhead costs.

(a) Bruce Matthews, Manager of the Advanced Fuels Section, Liquid Metal Fast Breeder Reactor program, Los Alamos Plutonium Laboratory, Los Alamos, New Mexico. Personal Communication.

TABLE 1. Capital Equipment Costs for Pellet Fabrication

Equipment/Procedure	Description	Manufacturer	Cost
2 Glove boxes	Inside floor dimensions: 5' 3" x 4' 11" 16 glove ports Box wall: 0.25" lead sandwiched between stainless steel sheets sheets 0.125" Windows: Leaded glass Gloves: Lead-loaded neoprene, 0.040" thick	Molitar Englewood, Colorado	$ 52,000
2 Balances	Cat. #3330-04 Load cell with remote controls and readouts. Dual range: To 3 kg, 0.1 g sensitivity; to 300 g, 0.01 g sensitivity	Scientech Boulder, Colorado	$ 4,100
Dry Granulator	ERWEKA Granulator Drive AR 400 Granulator TG 2/S	Chemical and Pharmaceutical Co., Inc. 225 Broadway, New York	$ 3,600
Blender	"Turbula:® Type T2C	Chemical and Pharmaceutical Co., Inc. 225 Broadway, New York	$ 3,000
Press	30 Ton Hydraulic, double acting Reservoir and pumps remote (outside glove box) All controls outside glove box	Western Sintering Richland, Washington	$110,000
Glove box installation	$10,000/box Engineering and Crafts: 425 h at $47/h		$ 20,000
Equipment installation	Press: 200 h at $46/h Other: 120 h at $46/h		$ 14,720
TOTAL			$207,420

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TABLE 2. Capital Equipment Costs for Powder Reconstitution

Equipment/Procedure	Description	Manufacturer	Cost
2 Glove boxes	Inside floor dimensions: 5' 3" x 4' 11" 16 Glove ports Box wall: 0.25" lead sandwiched between stainless steel sheets 0.125" Windows: Leaded glass Gloves: Lead-loaded neoprene, 0.040" thick	Molitar Englewood, Colorado	$52,000
Balance	Cat. #3330-04 Load cell with remote controls and and readouts. Dual range: To 3 kg, 0.1 g sensitivity; to 300 g, 0.01 g sensitivity	Scientech Boulder, Colorado	$ 2,100
Dry Granulator	ERWEKA Granulator Drive AR 400 Grnaulator TG 2/S	Chemical Pharmaceutical Co., Inc. 225 Broadway, New York	$ 3,600
Furnace	Model #51442 Control model #59344 (remote) 4800 watts Exterior dimensions: 20" W x 20" H x 24.5" L	Lindberg Watertown, Wisconsin	$ 1,950
Mill rack and mills	Rack Model #764AV: 30 1/4" x 12 3/4" x 15 3/4" H 3 Mills: Rubber-lined steel size 1 Stainless steel balls, 0.5", 100 lbs	E. T. Horn La Mirada, California	$ 2,310
Glove box installation	$10,000/box Engineering and Crafts: 425 h at $47/hr		$20,000
Equipment installation	160 hr at $46/h		$ 7,360
TOTAL			$89,320

TABLE 3. Start-Up Operation Costs

Process	Personnel	Job Description	Cost
Pellet fabrication	Engineer	120 h at $65/h Prepare detailed operating procedures in conjunction with an operator. Supervise equipment shakedown.	$16,400
	Operator	120 h at $50/h Operate equipment start-up and shakedown	
	---	Preparation of criticality specification: 40 h at $65/h	
	---	Radiation monitoring: Included in labor contract	
Powder reconstitution	Engineer	120 h at $65/hr Prepare detailed operating procedures in conjunction with an operator. Supervise equipment shakedown.	$16,400
	Operator	120 h at $50/h Operate equipment start-up and shakedown	

TABLE 4. Process Operation Costs

Process		
Pellet Fabrication	Estimate assumes 3 shifts/day processing a 100-kg minimum lot of PuO_2 powder.	
	Two operators/shift at $50/h/operator	
	Maximum 20 kg powder processed/day	
	Labor cost/kg	$120.00
	Radiation monitoring: Included in labor overhead.	
	Supplies/kg: Does not include items required for shipping as powder. Includes such items as stainless steel cylinders, neoprene lead-loaded gloves for replacement, organics.	1.50
	Only utilities: Electricity/kg	0.80 kWh
	Total pellet fabrication price/kg	$122.00
Powder Reconstitution	One operator/shift for 4 h at $50/hr	
	10 kg pellets processed to powder in 4 shifts	16 h labor
	Labor cost/kg	$ 80.00
	Radiation monitoring: Included in labor overhead.	
	Supplies/kg	$ 0.75
	Only utilities: Electricity/kg	12.0 kWh
	Total powder reconstitution price/kg	$ 81.00

TABLE 5. Cost Summary

Pellet Fabrication	
Capital Equipment	$207,420
Start-Up Costs	$16,400
Process Operation	$122/kg
Powder Reconstitution	
Capital Equipment	$89,320
Start-Up Costs	$16,400
Process Operation	$81/kg

Indirect Cost: Personnel Exposure

For these calculations, we have considered only the additional dose to workers (over usual doses) when processing unfired pellets for shipping and reducing them back to powder form for further processing. Any changes as a result of alteration in such matters as shipping container loadings were not considered. The calculational methodology used in Section 4 of BNWL-2086, A Guide to Good Practices at Plutonium Facilities (Faust et al. 1977), was used for estimating neutron and gamma dose equivalents. The shielding code PUSHLD (Strode and Van Tuyl 1973) was used to calculate the photon dose rate from cans of powder and pellets, since the complex spectra from plutonium make it impossible to use analytical functions for estimating photon shielding. The PUSHLD code has been used extensively in the past and has been experimentally verified with exposures made with high-exposure plutonium.

Plutonium emits photons (gammas and X-rays) as well as neutrons from spontaneous fission of even-numbered isotopes and alpha-neutron reactions with low-atomic-number elements in contact with the plutonium. In general, the photon dose predominates where there is little shielding, (e.g., a glove), and the neutron dose predominates where there is shielding for gamma rays (e.g., the steel and lead shielding found on glove boxes, which is easily penetrated by the neutrons).

For these calculations, it was assumed that the glove boxes used were patterned after a Los Alamos National Laboratory design. The glove box layout used for calculations is shown in the "Direct Costs" section (Figure 1 for the pellet fabrication and Figure 2 for the reduction of pellets back to powder). The glove boxes were assumed to be fabricated from stainless steel 1/4-in. thick, with an additional 1/4 in. of lead shielding. Also, all the windows were covered with leaded glass. Many different types of rubber gloves are

available for glove boxes. For these calculations; 0.040-in.-thick, lead-loaded neoprene gloves were used, since data for these gloves are available in the PUSHLD code library.

It was also assumed that there was no significant contribution to dose from other glove boxes nearby or from material stored in containers in the same room. This might not be the case, however, in many existing facilities where the pellet-processing glove boxes may be crowded into an existing facility.

The dose equivalents to workers processing plutonium are highly variable. The contact or hand exposures are especially difficult to estimate. The contact doses depend upon the skill of the operator (speed in completing a job), the operator's work habits, and adherence to ALARA (as low as reasonably achievable) philosophy for reducing dose. The following calculations were based on the plutonium-handling practices outlined in BNWL-2086 (Faust et al. 1977) and minimize the time during which quantities of plutonium dioxide are held in the hand. It was further assumed that at least four hours per week are spent cleaning up the glove boxes and that gloves are changed with reasonable frequency. Experience in handling high-exposure PuO_2 powder for fabricating fast breeder reactor fuel pellets has shown that a "background" of greater than 20 mrem/hr can result from dust layers on gloves and glove box surfaces. Glove boxes must be cleaned regularly to prevent excessive exposures. The contact dose equivalents could be increased by a factor of two if the operator has poor work habits; there could be a similar reduction if the process were more automated and fixtures were provided so that the worker's hand seldom contacted the PuO_2. For instance, forceps or tongs could be used to remove pellets from the press, to weigh them and to load shipping tubes. For the calculations presented here, it was assumed that the worker directly handles quantities of plutonium for 10 to 15 seconds for each transfer operation. For a highly skilled worker, however, this figure may be conservative.

External whole-body dose equivalents are not as variable as hand exposures, and are not as dependent upon the worker's habits. It was assumed that the glove boxes contain an additional 1/4-inch lead shielding and that all glove parts are covered with shielding to prevent radiation streaming out the glove parts. Neutron and gamma dose equivalent rates were calculated at 35 cm from the PuO_2 source; this is a convenient working distance for handling materials in a glove box. It was also assumed that some material was temporarily stored in the glove box at a distance of one meter from the operator or the health physics technician monitoring the operation.

Dose equivalent rates from plutonium sources vary, not only with shielding, but also with isotopic composition and with time. The time variation is due primarily to the growth of ^{241}Am from the beta decay of ^{241}Pu. For these calculations, it was arbitrarily decided to use one-year-old plutonium (i.e. plutonium chemically separated for one year), since it was felt that this is a conservative upper limit on the time required to separate, process, ship, then reconstitute the PuO_2 back to powder form. Since dose rates vary with isotopic composition, two extreme cases were considered to demonstrate the variation which can exist. The first, low-exposure plutonium, is somewhat typical of the material used in PuBe neutron sources in the past. The second is high-exposure

plutonium, which is the projected average isotopic composition for all the plutonium to be produced by light water reactors in the United States in the year 1985 (Faust et al. 1977). These two isotopic compositions are given in Table 6; the low-exposure plutonium is designated as low exp., and the high-exposure plutonium is designated as 85 LWR.

Based on these isotopic compositions and the data in BNWL-2086 (Faust et al. 1977), the low-exposure plutonium is shown to emit 126 neutron/second-gram of PuO_2, and the high-exposure 1985 LWR plutonium is shown to emit 650 neutrons/second-gram of PuO_2. Calculations of neutron dose equivalent rates were based upon these neutron yields. The effects of source multiplication were ignored; they are not significant for 1-kg batches of loose PuO_2 powder.

The processing of PuO_2 powder into unfired pellets for shipping and the reconstitution of the unfired pellets back to PuO_2 powder is given in the scenarios presented in Tables 7 and 8, respectively. Photon dose rates were calculated using the computer code PUSHLD (Strode et al. 1973), with the source sizes and densities outlined in the scenarios. Neutron dose equivalent rates were calculated using the methods in Section 4 of BNWL-2086 (Faust et al. 1977).

TABLE 6. Isotopic Compositions of Plutonium Used for Dose Calculations

Material	^{236}Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Low-Exposure (Low-Exp.)	---	0.128	93.7	5.81	0.139	0.03
Average of Pu Produced in Light Water Reactors during 1985 (85 LWR)	7.3×10^{-6}	1.60	54.7	24.7	12.2	6.70

TABLE 7. Scenario for Processing PuO_2 Powder into Unfired Pellets for Shipment(a)

1. Introduce the PuO_2 powder into glove box and weigh. It is assumed that the PuO_2 is a loose powder in a container 3.5 inches in height and diameter.

2. Transfer the weighed powder into the pellet press glove box.

3. "Slug" the powder in the pellet press to form low-density pellets 1.5 inches in diameter by 0.5 to 0.4 inches. The slugs are 1 kg of PuO_2 and are handled in 10 mil steel cans.

4. Transfer the slugs back to the glove box containing the granulator.

5. Granulate the "slugged" pellets to a coarse powder form.

6. Weigh the PuO_2 powder and binder and combine them.

7. Blend the PuO_2 powder and binder in batches containing 1 kg of PuO_2.

8. Transfer the mixture back to the pellet press glove box in steel cans.

9. Press the PuO_2-binder mixture into unfired pellets. The pellets are about 1.5 inches in diameter and 0.87 inch high.

10. Weigh the pellets and load 4 pellets into a stainless steel tube and bag out. It is assumed that the unfired pellets have a density of 5.8 g/cm^3 and are shielded by the stainless steel tubes with a wall thickness of 0.060 inches. Three tubes are loaded into a steel can containing about 1.75 kg of PuO_2.

(a) This process requires 2 operators and 1 health physics technician.

TABLE 8. Scenario for Reconstituting Unfired PuO_2 Pellets Back into a Usable Powder Form[a]

1. Receive the pellets, unload from the stainless steel tubes and weigh. It is assumed that there is about 1.75 kg of PuO_2 per can.

2. Granulate the pellets to a coarse powder form in a Erweka® dry granulator. It is assumed that the powder is placed into steel cans with walls 10 mil thick.

3. Transfer the granulated PuO_2 powder to the furnace glove box.

4. After 5 kg of PuO_2 are accumulated, load the granulated powder into a Lindberg[b] furnace and calcine in air for 12 hours to remove the binder.

5. Transfer the 5 kg of calcined PuO_2 back to the granulator glove box.

6. Ball mill the powder in 2.5 kg batches.

7. Unload the ball mill and weigh out 1 kg of powder into 3.5 in. x 3.5 in. diameter steel cans for processing or storage.

(a) This process requires 1 operator and 1 health physics technician.

(b) Lindberg Company, Watertown, Wisconsin.

® Erweka is a trademark of Chemical and Pharmaceutical Industry Company, Inc., New York.

Using times estimated from the processing scenarios, the dose equivalents for processing 5-kg batches of PuO_2 were calculated considering the following points.

1. Contact doses were calculated based upon 10-second hand contact times for directly handling containers of PuO_2 powder or pellets. It was assumed that lead-loaded neoprene gloves 0.040 in. thick, were used to handle the PuO_2.

2. Whole-body dose equivalents were calculated using four different sources. The most important contribution is from the material being handled by the operator. For calculational purposes it was assumed that the PuO_2 was 35 cm from the operator's body.

The second contribution is from other PuO_2, which is being temporarily stored in the glove box until a 5-kg batch is accumulated for further processing. It was assumed that this material was at a distance of 1 meter from the workers.

The third contribution is from other sources in the room. This includes gamma doses from dust layers on the inside of gloves which have been pulled outside the glove box so they will not interfere with operating machinery. These "background" exposures were estimated from unpublished data accumulated by PNL personnel for the operation of a ball mill, blender, and pellet press used to process high-exposure plutonium dioxide powder. Thermoluminescent dosimeters were used to estimate the average exposure rates over a four-month period during which the glove boxes were in use. When the gloves were not in actual use, a 1/4-in.-thick lead shield was placed over the glove parts to prevent excessive personnel exposure. For calculational purposes, the dose rates for low-exposure plutonium are estimated from the ratio of surface dose rates through the 0.040-in.-thick, lead-loaded neoprene glove for low exposure and the 1985 LWR plutonium described in Table 7.

The fourth contribution is from glove box clean-up operations, which are assumed to take four hours per week of processing operations.

The average dose rates from the study described above were used to estimate doses from cleanup operations.

The results of these calculations are presented in Tables 9 and 10 for processing 100 kg of PuO_2. Table 9 summarizes the estimated dose equivalent for a three-person crew fabricating unfired pellets using the scenario in Table 7. Two operators are required full time, and a health physics technician is required about one-fourth of the time. The health physics technician is assumed to be one meter from the sources and never handles them. The contact or hand dose considers only gamma exposures. It is very difficult to accurately calculate the neutron dose to the hand, and there is some uncertainty in applying a quality factor for neutrons based upon the induction of leukemia, since there are no blood-forming organs in the hands.

Table 10 summarizes the estimated dose equivalent for two workers processing the unfired pellets back into PuO_2 powder. It is assumed that the health physics technician is required only one-fourth of the time for bag-out operations, routine surveys, etc. and remains at one meter from the cans of PuO_2. The whole-body dose equivalents are lower in the powder reconstitution process because the process is somewhat more automated and the single operator spends less time with hands in the glove box.

The range values given in Tables 9 and 10 are estimated from the experimentally observed values of exposure rates described previously for high-exposure plutonium. Wide variations are possible between the dose equivalents

TABLE 9. Summary of Dose Equivalent Estimates for Fabricating PuO_2 Powder to Unfired Pellets

	Total Dose Equivalent for Three-Person Crew Processing 100 kg of PuO_2 (man-rem)	
	Average of Light Water Reactor Plutonium Produced in 1985	Low-Exposure Plutonium
Contact or hand exposure (gamma only)	67.0	18.0
Whole body dose equivalent including room background		
Average	0.95	0.14
Range based on variations in room background	(0.87 to 1.1)	(0.11 to 0.15)

TABLE 10. Summary of Dose Equivalent Estimates for Reconstituting Unfired PuO_2 Pellets Back to Powder

	Total Dose Equivalent for Two-Person Crew Processing 100 kg of PuO_2 (man-rem)	
	Average of Light Water Reactor Plutonium Produced in 1985	Low-Exposure Plutonium
Contact or hand exposure (gamma only)	64.0	17.0
Whole-body dose equivalent including room background		
Average	0.19	0.038
Range based on variations in room background	(0.14 to .26)	(.03 to .06)

calculated and those which could be experienced in actual processing. The calculations assume highly skilled personnel who adhere to ALARA dose philosophy. Small reductions in the whole-body doses could be achieved by automation, but fairly significant reductions in hand exposure could be achieved by almost eliminating direct contact with containers holding PuO_2. The dose equivalents could be considerably larger for unskilled operators or those with "poor" work habits -- allowing quantities of material to accumulate in the glove box, on the windows and gloves, not covering the glove ports when not in

use, allowing gloves to hang outside the glove boxes. No attempt was made to quantify these practices, but it is estimated that the doses could increase by well over an order of magnitude.

One other point should be considered, although it is outside the scope of this project. In the past there have been problems caused by the radiolytic decomposition of plutonium nitrate solutions from the intense alpha activity of plutonium. The effects of alpha radiation on the organic binder should be thoroughly investigated. Quantities of hydrogen gas could be liberated to cause pressurization or explosive hazards. It might be necessary to place constraints upon the time and/or the specific activity of the plutonium being shipped as pellets. High-exposure plutonium, with much higher specific activity from the ^{238}Pu and ^{241}Am isotopes, is expected to present much more of a problem than that of lower activity.

CRITICALITY ASPECTS OF ALTERNATE PLUTONIUM SHIPPING FORMS

Changing the shipping form of PuO_2 from an oxide powder to an unfired PuO_2 pellet would not significantly impact criticality safety for either shipping or handling. The nuclear safety limits generally used are based on theoretical-density material, so increasing the bulk density of the oxide from nearly 2 g/cm^3 to 5-6 g/cm^3 would still be considered safe. With considerable additional analysis, it might be possible to increase the allowable plutonium loading in shipping containers from that currently used. However, strong experimental evidence for the pellet behavior under accident conditions would be required.

Handling PuO_2

The basis for criticality safety is the double contingency criterion. This criterion states that at least two unlikely, independent, and concurrent accidents, failures, or limit violations must occur before criticality is possible. A standard factor of 45% of the critical mass limit is applied to meet the double contingency criterion when mass batch limits are involved. This limit will ensure that a double batch (assuming a double batch to be the maximum credible mass limit violation) will remain subcritical.

Figure 3 (Hansen and Clayton 1967) shows the water-reflected spherical critical mass for PuO_2-water mixtures with initial densities of 11.46 g/cm^3 and 5.6 g/cm^3. For dry mixtures (H/Pu$\leq$8), the minimum critical mass for theoretical density (TD) oxide (11.46 g/cm^3) is greater than 12 kg PuO_2. This results in a batch limit of 5.4 kg PuO_2 for TD oxide under the double contingency criterion. The only additional limit required is the exclusion of water. For uncontrolled geometries, the addition of water to a 5.4-kg batch could result in criticality, since the minimum critical mass for dilute PuO_2-water mixtures is only 531 g of plutonium (Carter et al. 1969). Thus, meeting the double contingency criterion requires either that it be incredible to moderate the dry PuO_2 or that additional geometry controls be applied to ensure that moderation alone will not result in criticality.

For PuO_2 with an initial concentration of 50% TD, the critical mass shown in Figure 3 is higher than for TD oxide. This increase is particularly large at low plutonium-to-hydrogen ratios. For this material, continuing to assume a 5.4-kg PuO_2 limit will certainly be conservative, resulting in a reduced fraction of critical mass. With sufficient experimental evidence of unfired pellet oxide density, it might be possible to raise this limit, if needed.

Transporting PuO_2

Many different containers have been certified for use in the transportation of plutonium oxide. The most common containers are listed in Table 11 along with the current shipping limits. Figures 4 through 8 (WASH 1279) show construction details for these containers. One study (Heaberlin 1978) indicated that most plutonium oxide shipped between DOE sites in 1976 was shipped in 6M or LLD-1 containers. For dry plutonium compounds, no limits are placed on the plutonium density. The criticality analyses used in certifying these containers assumed that density which results in the highest k_{eff} for the

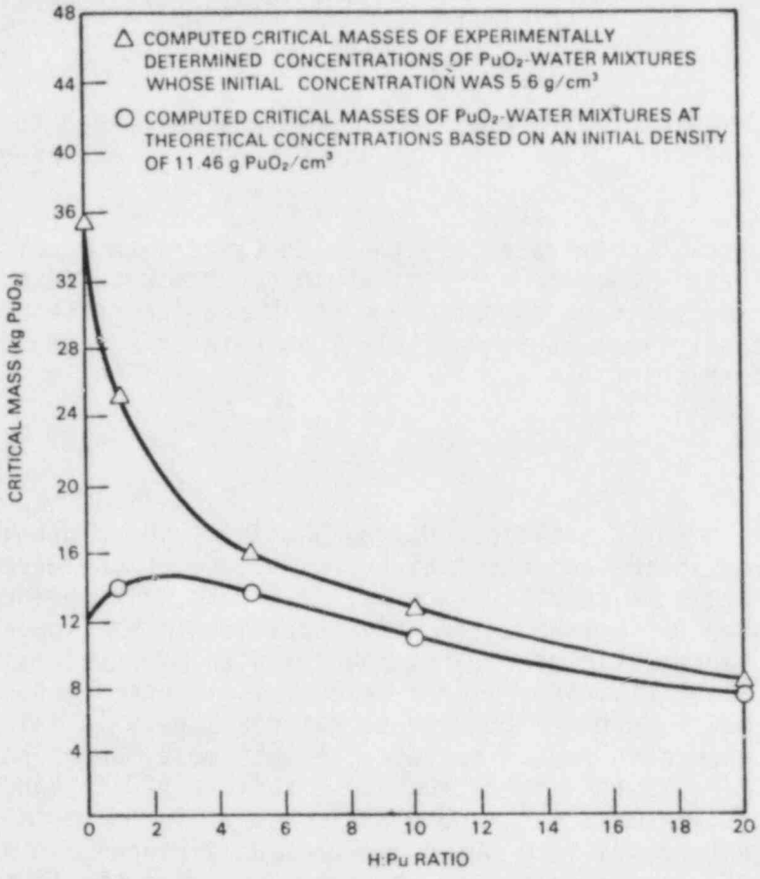

FIGURE 3. Water-Reflected Spherical Critical Mass for Experimental and Theoretical Concentrations of Homogeneous PuO_2-Water Mixtures (Hansen and Clayton 1967)

TABLE 11. Limits for PuO_2 Shipping Containers

Container Type	PuO_2 Mass Limit[a]	Moderation Limit
6M (Spec.)	4.5 kg[b]	H/Pu $\leq$ 3
LLD-1	4.5 kg	
6L (Spec.)	2.5 kg	H/Pu $\leq$ 10
5795	See below[c]	
L-10	4.5 kg	Dry

(a) Mass limits based upon criticality analysis. Radiation level or decay heat rate may be limiting.
(b) 5 wt% ^{240}Pu minimum.
(c) For the 5795 Foamglass container, the maximum plutonium limits are 16.5 kg oxide for H/Pu $\leq$ 0.4 (8.73 g Pu/cm^3) or 18.3 kg oxide for H/Pu $\leq$ 3 (4.71 g Pu/cm^3). The plutonium must contain at least 60 wt% ^{239}Pu, and more ^{240}Pu than ^{241}Pu; ^{238}Pu shall be considered part of the ^{239}Pu.

container array. Therefore, no additional analysis is needed to allow transport of the unfired pellets in this study under the existing container limits.

It might be possible to recertify these containers to specifically ship large unfired pellets. Such an analysis would require knowledge of the effects of different assumed accident scenarios on the container contents, possibly also a thermal reanalysis. It is unlikely that this effort would result in greatly increased shipping limits.

BENEFITS

The principal benefit anticipated from modifying the plutonium shipping form is a reduction in the potential airborne release of the material under off-standard and accident conditions during transport. The principal hazard for insoluble plutonium is inhalation and deposition in the lungs. Therefore, the important characteristic of PuO_2 is the fraction of particles 10 μm Aerodynamic Equivalent Diameter[a] or less that is injected into a flowing gas stream (aerosolized). In order to release materials held in shipping containers and make them airborne, a series of events must occur and a number of conditions satisfied. First, there must be a failure of all barriers--high-strength containers, product can, and plastic bags. The material released must be in--or converted into--a form which can be made airborne. A significant fraction of a powder can be made airborne depending upon the level and type of

(a) Particles which exhibit aerodynamic behavior equivalent to a sphere of density 1 of the stated size.

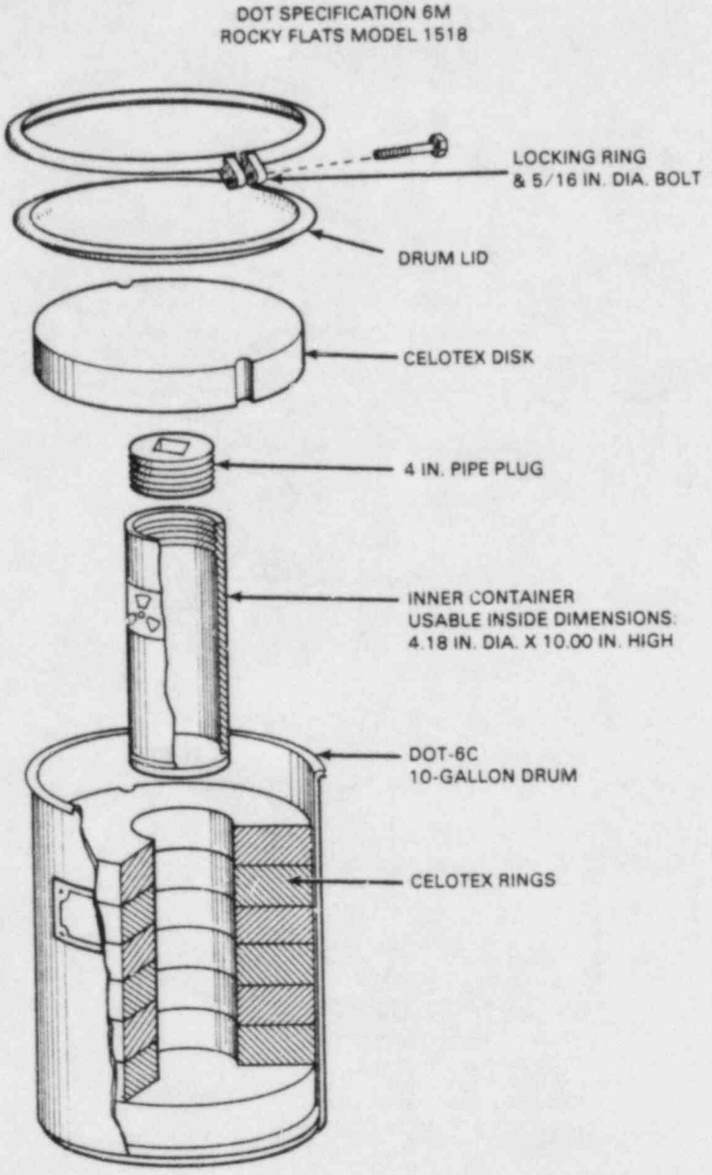

FIGURE 4. DOT Spec. 6M Container (after WASH 1279)

entraining force applied, but pellets must be converted. Short of catastrophic loss of all barriers, the size and characteristics of the leak and the level and type of force expelling the material also influence the amount and characteristics of the release. Whether the material is made airborne in the container and subsequently released, or is released (bulk release) and subsequently made airborne influences the type and level of entraining forces to which the material can be subjected.

The characteristics of pellets would reduce their potential for airborne release. First, there normally are essentially no particles present. If a force is applied which can subdivide a material and make it airborne, all that portion of the force which can normally be applied is available to entrain the

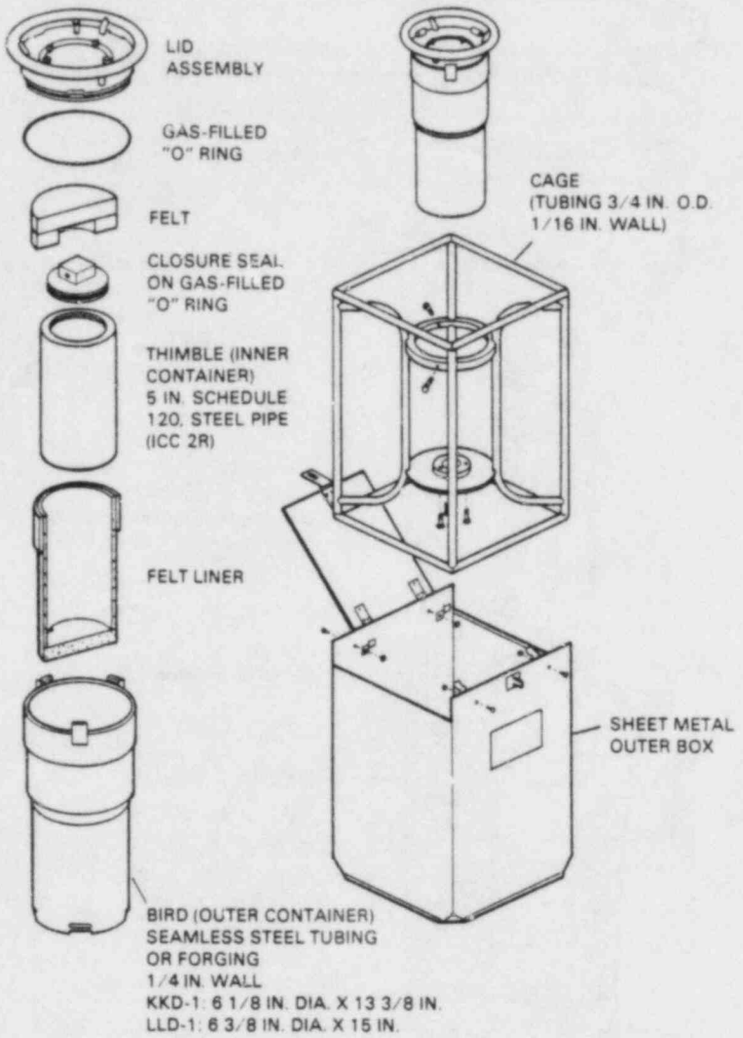

FIGURE 5. LLD-1 Container (after WASH 1279)

powder; however, a portion must be used to subdivide a pellet, reducing that available to propel the fragments. Powders can be released through smaller openings than most pellets, although fine powders like the PuO_2 considered in this study do not flow readily. Inasmuch as data is lacking (Panesko 1983) on the behavior of unfired pellets to various stresses and the number of possible situations involved, precise numerical evaluation of the reduction in dispersibility is not possible at this time. The comparison will be made upon the inferred behavior of unfired pellets and powder under the set of accident conditions covered in NUREG-0170 (NRC 1977, Vol. 1).

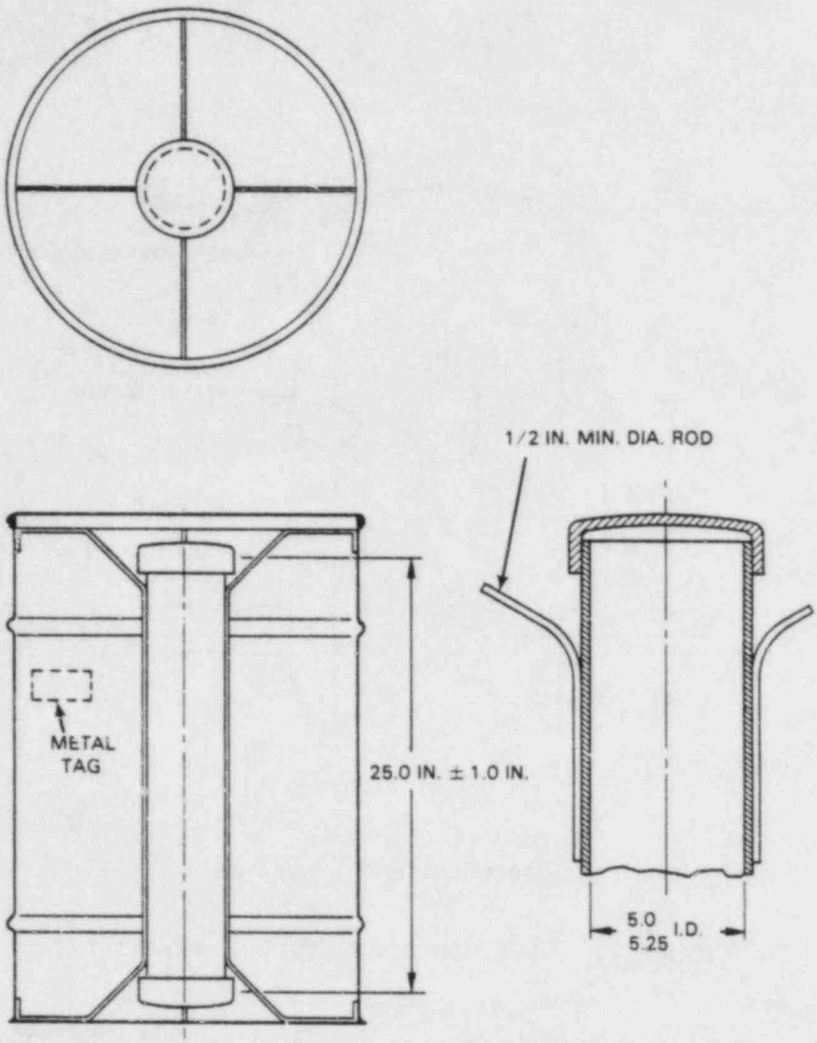

FIGURE 6. Spec. 6L Container (after WASH 1279)

Transport Accident Scenario

Transportation accidents are divided into eight severity classes for each mode of transport in NUREG-0170 (NRC 1977, Vol. 1). Severity classes range from I through VIII with increasing potential damaging stress. Various combinations of stresses (i.e., impact-fire, crush-fire, impact/puncture-fire), are used to define the ranges of severity for each mode of transport. Transport containers are considered by choosing values for severity classes to reflect anticipated damage (e.g., a combination of impact-fire is used to catagorize severity classes for aircraft and train, but severity class VI, where containers lose integrity, is at an impact velocity range of 224 to 304 kph for aircraft and a puncture velocity of 97 to 130 kph for trains).

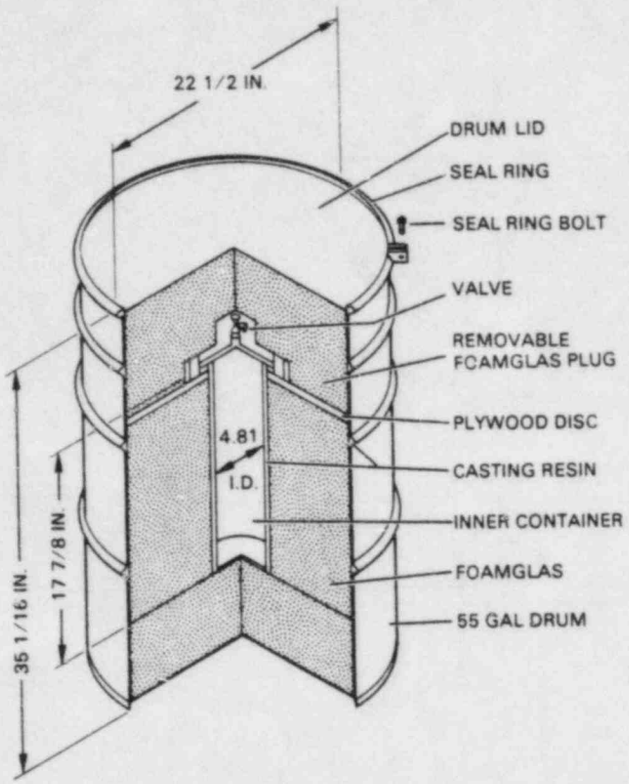

FIGURE 7. 5795 Container (after WASH 1279)

Quantities of contained materials released are assumed to be 1%, 5%, and 10% for the three upper-severity categories VI-VIII, respectively. The plutonium dioxide powder shipped is postulated to have a size distribution which has 20% of the material in the "respirable size (assumed to be 10 μm AED or less) range." A quarter (5%) is assumed to be made airborne by accident-generated/associated conditions.

The information appears to only address only one of two possible accident sequences: compromise of the containers--explusion of some of the material from the container and aerosolization of part of the released material by external forces. The second possible sequence appears to be aerosolization by accident-generated internal forces, compromise of the container, expulsion of a fraction of the airborne and bulk powder, and aerosolization of a fraction of the bulk powder released. The sequence followed apparently depends upon the accident considered and the type of force compromising the container. Crush can result in the catastrophic rupturing of a container that aerosolizes and releases the powder. Impact/puncture can result in various types and sizes of openings and can also aerosolize the powder. Fire can generate internal

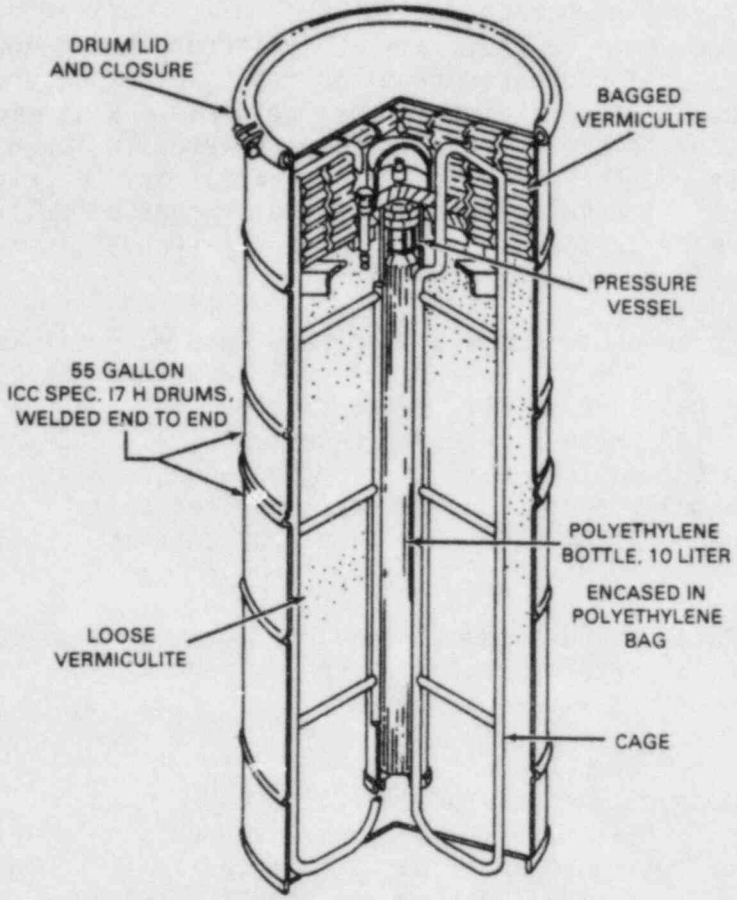

FIGURE 8. L-10 Container (after WASH 1279)

pressures, resulting in minute airborne releases for small openings (Owczarski et al. 1980) and large airborne releases from rapid depressurizations (Sutter 1983). The respirable fractions airborne (NUREG-0170, NRC 1977, Vol. 1) appear to be the following:

- class VI - $0.01 \times 0.05 = 5.4 \times 10^{-4}$ contents
- class VII - $0.05 \times 0.05 = 2.5 \times 10^{-3}$
- class VIII - $0.1 \times 0.05 = 5 \times 10^{-3}$

Dispersibility of Unfired Pellets

It is difficult to quantify the amount of material dispersed from unfired pellets as a result of various postulated accidents. First, the fraction of the force imposed upon the containers transferred to the contents is not known. Secondly, even if the forces imposed upon the contents could be defined, there is no information on the response of the unfired pellets to various levels and types of forces. Finally, there is no information on the responses of the various internal barriers to the forces applied and only limited data on the high-strength container, so the size and configuration of the leak path to the biosphere is another unknown.

For the purposes of comparison of the two forms (unfired pellets and powders), the assumption was made that, all other factors being equal, the fractional airborne release anticipated from the two types of materials is directly related to the fraction of particles in the "respirable size range." This approach does not address the release fraction covered in NUREG-0170 (NRC 1977, Vol. 1). It was assumed that although less material may be released from the container due to the relative coarseness of the fragmented pellets, a proportional quantity of material in "the respirable size range" is released and suspended under the same accident conditions.

The question to be addressed is, then, how much material in the "respirable size fraction" is formed from unfired pellets under accident conditions? Unfired pellets are relatively soft. Most studies indicated reconstituting the powder by hand grinding through screens (Panesko 1983). But these phenomena are associated with the pellets used in slugging and granulating processes and are not the unfired pellets considered here. Unfired pellets are pressed at higher pressures and only lack the high-temperature treatment to become fuel pellets.

Data are available on the impact behavior of sintered pellets (Panesko 1983). Sintered pellets were impacted at speeds ranging from 50-75 m/s (180 to 280 kph). The fraction less than 10 μm

- increased with speed
- decreased with temperature
- decreased with density of the pellet
- decreased with initial size (pellets vs. fragments).

The largest fraction less than 10 μm was generated from fragments impacted at 76 m/s with values around 7%. The size measurement technique was not given; therefore, it is not known whether the size is AED, least linear diameter, or another. If it is least linear diameter, the AED could be as much as 33 μm, considerably reducing the fraction less than 10 μm AED (respirable size). The fractions less than 10 μm for low-density pellets were generally less than 1% (0.29, 0.42, 0.6 and 2.85%).

In another series, sintered pellets were impacted at 58 m/sec (130 mph) at temperatures of 820°C or 850°C . The fraction less than 33 μm AED ranged from undetectable to 0.4%, which is roughly comparable to the study mentioned above. For sintered pellets, ultimate strength increases with sintering temperature. The pellets fail through brittle fracture, although the behavior of the pellets (as opposed to fragments) indicates that the consolidation may be greater on their surfaces than within them. Since the size distribution of the powder used to form the pellets is not known, the degree to which the forces applied reconstituted the size distribution of the starting material cannot be estimated. Subdividing the original powder required hours of milling.

For unfired pellets, the comminution mechanism would not be brittle fracturing but overcoming the adhesive forces holding the pellet together. Separation would occur where the forces were least effective. Whether this would occur at the individual particle level or between aggregates is not known. The capacity for packing particles together depends, among other factors, upon the size range and shape of the particles involved. Fine powders such as the PuO_2

used in fuel fabrication pack well under normal conditions just by settling and do not flow well. Therefore, it is assumed that force applied over a considerable time (e.g., hours) may be required to reconstitute the original size distribution.

The impact velocities used in the two studies are well beyond the speed considered (with no fire) of the highest severity class for trains. The impact speeds used in the study (110 to 195 mph) far exceed the speed anticipated for this mode of travel, and therefore exceed even more the speed the pellets may attain as a result of an accident. If we assume that the size distribution of the starting powder was 100% less than 10 μm (the information in Panesko 1983 seems to indicate that this type of size distribution was desirable for the starting material), as much as 7% could be returned to its original size. If the starting material were 20% less than 10 μm AED, 1.4% of the particles produced would be less than 33 μm AED, and somewhat less than 1.4% would be in the "respirable size fraction." Based upon the impact studies and the uncertainties in measurement units, a value of 1% is chosen for particles in the "respirable size range" for unfired pellets impacted at the velocities assigned to severity class VIII.

The response of unfired pellets to elevated temperatures does not appear to be comminution. Time is the variable fire parameter for severity classes, ranging from 0 to greater than 2 hours for 1300°K (1027°C). Higher temperatures (1300° to 1500°C) are used for sintering. Pellet strength increases with temperature; therefore, heating the unfired pellets should not result in any deleterious consequences.

Crush and fire are the stresses used to define the severity classes for motor vehicles (trucks). The effects of fires were discussed in the preceding paragraph. According to NUREG-0170 (NRC 1977, Vol. 1) the effects of crush appear to suggest a damage level to the high-strength container comparable to that inflicted by impact/puncture. This level of damage does not include a complete collapse of the high-strength container (where essentially 100% of the contents would be released). Under these conditions, where the high-strength container retains essentially its original shape, the enclosed materials would not be affected by the crush forces which are absorbed by the container. Any particle generation would result from the stresses imposed by normal handling and from the impact during the accident. It is assumed that the impact velocities are comparable to or less than these used to define the severity classes for trains; otherwise, the damage to the high-strength container would be greater. Thus, it is assumed that the fraction of particles in the "respirable size range" generated by these conditions is equal to or less than that generated in the same severity classes for trains.

There is no data on the behavior of sintered pellets at speeds comparable to those used to define the lesser severity classes for which releases are postulated (VI & VII). Although it can be stated that the degree of fragmentation is a function of the force applied, the actual relationship between the particle size distribution generated and the type and level of force applied is

not known. For the purposes of this analysis, it is sufficient to know that it will be less than that assumed to produce the severity class VIII releases.

Potential Reduction of Airborne Releases During Shipping Accidents by Use of Unfired PuO_2 Pellets

Based upon the availability of particles in the "respirable size range," use of unfired PuO_2 pellets as a shipping form would reduce the potential airborne release from a shipping accident by a least a factor of 20. Additional reductions may be affected by packaging practices used for the pellets. For instance, three stacks of 1.5-in. diameter x 3 (.87 in.) tall pellets could be included in a product can. Each stack of three pellets would be placed in a stainless steel sleeve and sealed in a polyethylene tube to facilitate handling and packing. A close-fitting metal sleeve should aid in reducing "dusting" during routine handling and particle generation under accident conditions. If a stack of pellets were bagged out of the enclosure using a polyethylene tube, canning would be a clean operation, and sealed cans would strengthen the internal barriers to release. Short of a catastrophic failure of the high-strength container, there is not a single set of accident conditions which would totally eliminate all internal barriers. Impact/puncture can cause the rupturing of thin plastic and metal holders. Material can leak from the compromised barriers; but their remnants, retaining much of their original shape, are still an impediment to flow, especially gravity flow. Elevated temperature can cause melting and vaporization of plastic and distortion of thin metal. Again, the remnants provide a impediment to flow. For sealed containers, elevated temperatures lead to increased pressure and can lead to compromise of the container. As mentioned before, crush is only a significant factor if the high-strength container is seriously distorted.

Leak size and configuration are also important considerations. At lower damage stress levels, it is anticipated that the leak paths are smaller and the fragments larger. Lesser fractions of the pellet fragments than powder would be released and pathways plugged more readily, exposing less-hazardous material to external forces.

It is anticipated for all of the above reasons that use of unfired pellets would significantly reduce the potential airborne release during accidents. Furthermore, the use of pellets rather than powder could result in a reduced potential for accident criticality due to flooding. Other benefits could include fewer material losses during transport (although total losses because of additional process steps might be greater), ease in the packing steps because of a more definite form (an inner container with less free volume could be used), and innovative packaging (utilizing the additional available space) to increase the reliability of the inner barrier.

EXPERIMENTAL PLAN

Although the technologies required for pelletization and reconstitution are available as interim steps in existing processes, some data essential to a full evaluation of the cost benefit of utilizing unfired pellets as an alternative plutonium shipping form were not uncovered during this and the previous

phases of the study (Mishima and Lindsey 1983). The essential data are in the areas of process parameters and pellet behavior, with packaging an optional area. The first two areas appear to be inseparable. The objective would be to generate pelletizing parameters to produce pellets that generate the smallest fraction of particles in the "respirable size range" that can readily be reconstituted into an acceptable starting material for fuel fabrication (that does not require requalification of the fuel pellets). No attempt was made to determine the costs for such an experimental program, although it may well represent the greatest direct cost of converting to an alternative plutonium shipping form.

Before an experimental program can be undertaken, some preliminary decisions are required concerning the size distribution and characteristics of the feed and reconstituted powders, the range of isotopic composition of the plutonium, shipping and handling procedures, and so on. Initially, pelletizing parameters would be studied, such as press pressure and binder type and quantity, in order to maximize some meaningful criteria of strength (such as percentage of theoretic density). The pellets generated would then be used for a limited study of response behavior under transport and accident conditions (size distribution of fragments/powder as a function of impact/puncture velocity, crush pressure, thermal shock,) and powder reconstitution parameters compared to the same strength criteria. Once the choice was made as to which particular mix of pellet strength, response behavior and reconstitution provides the most satisfactory blend of characteristics, a complete study of all three areas would be undertaken. Any study to optimize packaging techniques (to improve package integrity, to reduce internal pressure and "dusting" of pellets, etc.) could also begin at this time.

Pelletizing Parameters

The principal pelletizing parameters identified (Panesko 1983) are pretreatment, binders (including moisture) and pressing pressure. The information is sparse and directed towards producing strong fired pellets. Most of the measurements of the strength of the pellets (ultimate stress and strain) and their behavior have been performed on the final product, fired pellets. Data are required on the effects of binders and press pressure on unfired-pellet strength. The measure of pellet strength of interest in this study is resistance to fragmentation into particles in the respirable size range and (of lesser concern) "dusting" during normal handling and transport. Percentage of theoretical density is another measure of strength which is easier to obtain and can be used as a initial index for pellet evaluation. More information is available on pretreatment (ball milling of powders, prepressing, dry slugging and granulation); furthermore, the effects of powder characteristics on unfired strength can be alleviated by a judicious choice of specifications for starting materials.

Experimental studies are recommended to define the relationship between the percentage of theoretical density and each of the following parameters:

- press pressure
- binder type (organic compound moisture, etc.) at a specific press pressure
- quantity of a specific binder at a given press pressure.

The initial set of experiments, without binder, will define the optimum press pressure for unfired pellets. Visual observations will also be required to define pellet characteristics. Once the optimum press pressures are established, the efficacy of various binder types at a range of pressures around the optimum could be determined. If these tests indicated a serious divergence from the optimum pressures, the pressure range could be extended. The minimum mass of the most effective binders would then be tested over a range of pressures around the new optimum.

Reconstitution Parameters

Some of the pellets generated in the pelletizing tests, with and without binder, will be tested to determine the most effective techniques to regenerate a powder to a satisfactory starting material for fuel fabrication. Ball milling (along with heating for binder removal) was used in this study to estimate direct costs. This technique is currently in use to homogenize PuO_2 powders for starting material in fuel fabrication. Granulation was through a 35-mesh screen (Panesko 1983). Other techniques may be available which are more effective, and a thorough review for such techniques should be performed prior to the initiation of experimental work.

Once the technique or techniques for reconstituting the powders and the criteria for a satisfactory starting material for fuel fabrication have been chosen, an experimental program can be undertaken to determine the reconstitution process parameters. Some, but not necessarily all, of the process parameters of concern are defined below:

- How long, at what temperatures and under what atmosphere must the pellets be treated for binder removal? What are the effects on the process and product?
- What, if any, treatment is required prior to binder removal?
- If milling is the comminution process, what type of break-up materials (balls, rods, etc.) are used? For how long, under what conditions? What are the losses?

Response Behavior

Data are required on the quantity and size distribution of particles generated by unfired pellets under a variety of stresses arising during routine

transport or accidents. Stresses which may be encountered during routine transport are vibration and low-level impact. Accident-generated stresses are impact, crush, elevated temperatures, thermal shock and unusual environment (possible at elevated temperatures). Stresses may be encountered individually or in combination. Tests to evaluate the response behavior of unfired pellets should be conducted in the same manner (individually and in various combinations and sequences) with and without the stainless steel sleeve.

Pellets having a range of strengths (as measured by percentage of theoretical density) around the optimum value determined in the pelletizing and reconstitution process parameters study would be used in the study of response behavior. The following studies are recommended:

- size distribution of fragments (in Aerodynamic Equivalent Diameters) as a function of impact velocities from 20 fps (or minimum velocity to fragment pellets) to 450 or 500 fps (307 or 341 mph), with and without sleeve

- size distribution of fragments at crush forces from that required to break pellets to 330,000 lbf, with and without sleeve

- size distribution of fragments as a result of heating in the range of 200°C to 1300°C and cooling to ambient temperatures:

  - in air, cooling at a natural rate

  - in combustion products, cooling at a natural rate

  - in gaseous extinguishment agents (Halon®, CO_2, etc.), cooling at a natural rate

  - quenched rapidly in water.

- size distribution of fragments of heated and heated/cooled pellets above as a function of impact velocities and crush forces, with and without sleeve

- mass loss (and size distribution of fragments if possible) as a function of vibratory frequency and amplitude

- gases generated by radiolytical decomposition of organic binder if used.

The impact velocity study could be performed by impacting the object at the desired velocity against an unyielding stationary surface or against a massive, unyielding mass traveling at the desired velocity. The former technique is preferred since it allows greater control of the fragments. Crush force tests would use standard crush force presses with modifications to

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control the dispersion of fragments. Heating and cooling tests present no difficulty. A standard clam-shell-type tube furnace can be used for atmospheric heating and control. An adaptation to allow immersion of the heated object in a quench tank would be required. Equipment is available to control the frequency and amplitude of vibrations. A container could be fabricated to represent the space available for various configurations. Mass loss could be determined by weight differences over various periods of vibration. The fragments generated could be collected from the container and blown from the pellets by an air jet for size distribution measurements. Thus, techniques and equipment are available to conduct all of the outlined experiments.

Packaging Study

One optional study which appears to be of potential value is the investigation of various inner package components. Currently, packaging components within the high-strength container are several layers of plastic (generally PVC in the U.S.) and a sheet metal can. Closure of the can is by slip-fit and tape or by crimping. The use of unfired pellets with a known shape and rigid structure presents opportunities to further reduce the dispersion potential for the enclosed material under accident conditions.

For instance, if each stack of three unfired pellets could be bagged out of a glove box into a close-fitting, sealable plastic tube, the amount of plastic inside the can would be reduced. The bagged stack of pellets is "clean" and could be placed into a reusable stainless steel holder with slots to hold the stack. The holder would be designed to fit snugly into the can, which would then be crimped shut. Such an arrangement would reduce the quantity of plastic, thus reducing the internal driving force for dispersion in the event of a fire.

Other adaptations also suggest themselves:

- The tube used could be made of metal (e.g., aluminum or a low-melting alloy), eliminating the plastic entirely. Use of a metal tube increases the difficulty, during the "bagging-out" operation, of keeping its exterior surface uncontaminated.

- If a low-melting metal is used as the initial pellet container, the holder could also be made of the same material. Both would melt in the event of a fire and trap any fragments formed by the fire or other stresses. However, the potential for a physical explosion as a result of contact of the liquid low-melting with pools of water must be given serious consideration.

- The inner can could be equipped with a filtered pressure relief device to eliminate the driving force for fragment dispersion.

There are probably various innovative techniques that could be applied to each aspect of improving the containment capability of the inner packaging. A more systematic comprehensive survey would be beneficial.

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A limited cost-benefit evaluation was performed concerning use of unfired plutonium dioxide pellets as a shipping form. Two specific processing operations are required for this use, one to form the pellet and a second to reconstitute an acceptable powder upon receipt. The direct costs for the pelletizing process are approximately $208,000 for equipment and installation and $122 per kg of plutonium processed (based upon a 20-kg plutonium/day facility). The direct costs for reconstitution are approximately $90,000 for equipment and installation and $81 per kg of plutonium processed. The indirect cost considered was personnel exposure from these operations. Whole body exposures ranged from 0.04 man-rem per 100 kg of low-exposure plutonium reconstituted to 0.9 man-rem per 100 kg of average-exposure plutonium pelletized. The principal benefit is a potential twentyfold reduction of airborne release in event of an accident. An experimental plan is outlined to fill the data gaps uncovered during this study in the areas of pelletizing and reconstitution process parameters and pellet response behavior to accident-generated stresses. A study to enhance the containment potential of the inner packaging used during shipment is also outlined.

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