CONF-970126--9

LA-UR- 96-4766

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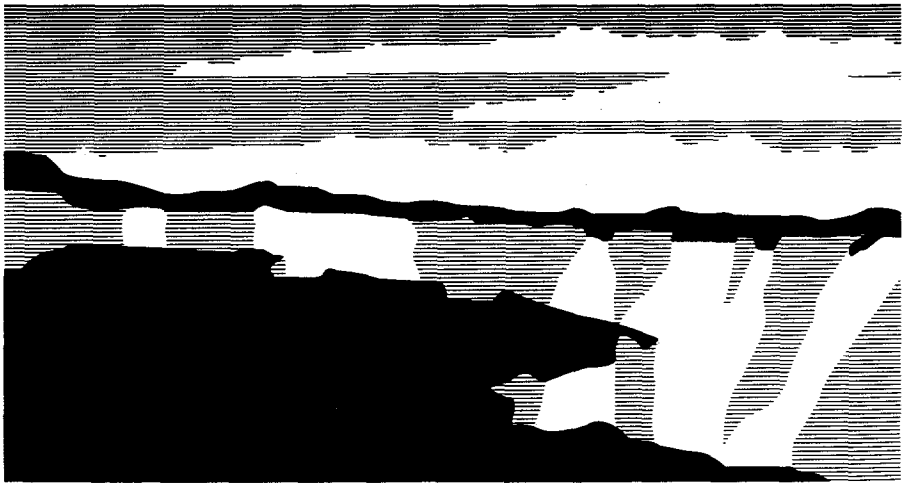
Submitted to: 5th Nondestrictive Assay & Nondestructive Examination Waste Characterization Conf., Salt Lake City, UT January 14-26, 1997



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NDA PDP PROGRAM PuO_2 INCREASED PARTICLE SIZE SPECIFICATION AND DESIGN

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ABSTRACT

Provisions in the National TRU Program Quality Assurance Program Plan require an assessment of performance for nondestructive waste assay (NDA) systems employed in the program. This requirement is in part fulfilled through the use of Performance Demonstration Programs. In order to optimize the quality and quantity of information acquired during a given Performance Demonstration Program cycle, the assessment employed is to be carefully specified and designed. The assessment must yield measurement system performance data meaningful with respect to NDA system capability to accommodate attributes of interest known to occur in actual waste forms. The design and specification of the increased particle size PuO_2 PDP working reference materials (WRMs) is directed at providing a straightforward mechanism to assess waste NDA system capability to account for biases introduced by large PuO_2 particles. The increased particle size PuO2 PDP WRM design addresses actual waste form attributes associated with PuO_2 particle size and distributions thereof, the issue of a known and stable WRM configuration and equally important appropriate certification and tractability considerations.

INTRODUCTION

Nondestructive waste assay methods are used in the National TRU Waste Characterization Program to determine the mass and associated alpha activity of waste entrained radionuclides. The capability and performance of waste NDA systems employed in the Program must comply with requirements as set forth in the TRU Waste Characterization Program Quality Assurance Program Plan (QAPP).[1] Compliance with QAPP and NDA requirements and objectives are in part assessed through the use of a Performance Demonstration Program (PDP). All waste characterization facilities that intend to ship TRU waste to the Waste Isolation Pilot Plant (WIPP) are required to participate in the NDA PDP. The NDA PDP Plan provides a means to institute a measurement routine to yield quantitative performance data for key NDA system parameters under specified conditions. The QAPP and NDA PDP, employed in conjunction, are designed to ensure that nondestructive waste assays performed to characterize the radionuclide content in containerized

TRU waste, produce data of known and defensible quality. The NDA PDP also allows a complex-wide assessment of NDA system performance relative to a common basis.

Implementation of the NDA PDP plan for the 55 gallon type container requires a carefully specified apparatus. The NDA PDP measurement apparatus consists of 55 gallon matrix drums and insertable radioactive standards. The PDP radioactive standard(s), hereafter referred to as working reference materials (WRMs), are designed to be configured with matrix drums in a particular manner to test system performance with respect to the objectives of a given test cycle. The Increased Particle Size (IPS) WRM set is specifically designed to allow NDA PDP program assessment of bias induced in waste NDA measurement systems by large PuO_2 particles, i.e. greater than 400 microns diameter, or the accumulations of numerous smaller particles known to exist in actual waste forms. The initial task in developing IPS PuO_2 WRMS is to determine a nominal increased PuO_2 particle size which yields and NDA system bias commensurate with that observed in measurement of actual waste configurations. Based on the specified particle size, fabrication and assembly techniques were developed at the Los Alamos National Laboratory to produce large particles of PuO_2 and WRMs possessing the attribute for use in the program. The specification, design and fabrication of IPS WRMs is addressed in this paper.

SPECIFICATION

The specification of a PuO_2 particle size which yields a bias representative of actual waste forms was determined through the evaluation of NDA system responses where bias resulting from increased particle sizes and/or accumulations of numerous smaller particles effectively inducing the same bias, is evident. For this purpose, the response of a commonly employed passive/active neutron NDA system to a large population of actual waste drum measurements, was evaluated. The approach for extracting the magnitude of the IPS bias representative of actual waste drum measurements, consisted of identifying a population of drums spanning several waste types and evaluating the depression in the active mode response relative to the passive mode response. This technique required several steps to filter out drums with bias and precision sources *not* due to increased PuO_2 particle size or agglomerations of numerous smaller particles.

The first step in quantifying the IPS bias source was to select several waste forms which have increased PuO_2 particle sizes by observation of real time radiography images or from information on the waste generation process. It was important that these selected waste types not have exceedingly convoluted bias and precision sources that complicate the extraction or bounding of the increased particle size bias effect. Waste types selected for assessing PuO_2 particle size effects consisted of graphite, three different combustibles classifications, and metals. Next it was

necessary to identify in this population drum assays where the passive neutron mode measurement not significantly impaired by known interferences. Drum assays exhibiting excessive moderator and absorber properties were filtered our, leaving approximately 3,000 passive/active neutron measurements from which to derive an estimate of a nominal IPS induced bias.

With this select population, the IPS bias was determined by taking the ratio of the passive mass assay to the active mass and averaging this parameter over the population. This ratio will in actuality reflect the complete range of fissile material physical configurations from diffuse distributions of Pu atoms to local accumulations of fine particles to large individual clumps of Pu —and any combination thereof—existing in the population under study. The range of the IPS bias parameter estimate, derived from the conditioned population, was 3.0 to 6.5. This range estimate was discussed with waste NDA individuals at other sites to ensure that the value was reasonable and reflected experience with waste forms at these facilities. They concurred with the range, and a value was selected from the range as the basis for determining PuO_2 particle sizes which would yield such a bias. It should be noted that it is not critical that an exact representation of the waste-form derived IPS value be determined; a range is sufficient. It is important that the PDP program include a test of NDA system capability to account for this bias source and that the magnitude of the bias represented in PDP assessment apparatus, i.e., WRMs, be reasonable.

Having determined a value of IPS bias, it is necessary to use this information to determine a particle size which would induce a bias of this magnitude for performance assessment purposes. The technique used for this purpose was Monte Carlo Neutron Photon (MCNP) modeling.[3] The approach consisted of computing the number of fission-induced neutrons produced by a thermal neutron interrogating spectrum (Maxwellian) incident on a PDP WRM model from all directions. The computations were performed for both homogeneous PuO_2/diatomaceous earth mixture and heterogeneous PuO_2 particle(s) distributed in diatomaceous earth cases. Several heterogeneous PuO_2 particle(s)/diatomaceous earth configurations were modeled to span the WRM-mass range of interest. To simplify modeling efforts, the particles were represented in a spherical geometry. The MCNP PDP WRM model consisted of the homogeneous mixtures and heterogeneous distributions for low burnup, weapons grade PuO_2 within the dual encapsulation cylinder specifications and dimensions of the standard PDP WRM.

An evaluation of the active mode, thermal-neutron, induced fission-yield depression due to increased PuO_2 particle sizes requires a baseline definition. The baseline for comparing an IPS-induced active-mode response depression is an "infinite dilute" case of PuO_2, 0.001 grams, distributed uniformly throughout the WRM diatomaceous earth diluent. To obtain baseline

response data, an MCNP model was constructed to the specifications of the actual PDP WRM encapsulation assembly containing the homogeneous mixture configuration. The number of thermal-neutron interrogation, induced-fission neutrons yielded from this model was then computed to give the baseline yield. Next, a model of the heterogeneous IPS WRM containing spherical PuO_2 particles distributed in diatomaceous earth was constructed. A number of heterogeneous cases were modeled and computed for WRMs through the mass range of 0.25—16.0 grams PuO_2 for three different spherical particle sizes.

Data generated from the set of homogeneous and heterogeneous MCNP WRM cases are plotted in Figure 1, illustrating the reduction in the number of fission-induced neutrons per PuO_2 mass as compared to the homogeneous "infinite dilute" baseline case. This ratio is referred to as the IPS induced self-shielding factor. As is evident from the plot, the ratio for the homogeneous case WRMs is nearly 1.0 at the low, 0.25 gram PuO_2 loading, and drops to approximately 0.75 at 16.0 grams. For the heterogeneous IPS cases, the same self-shielding factor was calculated over the 0.25—16.0 mass range for three different spherical PuO_2 particle sizes. For the 2,000 micron spherical particle heterogeneous case, the self-shielding factor is approximately 0.25 at 0.25 gram PuO_2 loading and remains essentially unchanged to the 16.0 gram mass loading. For the 2,500 and 3,000 micron spherical particle size case, the self-shielding factors are approximately 0.22 and 0.19 respectively, and are effectively unchanged over the 0.25—16.0 gram mass range. It is noted in the Figure 1 plot of self-shielding factors versus PuO_2 mass for the heterogeneous cases that the span over 2,000 to 3,000 micron particle sizes induces a self-shielding factor variation of approximately 6% at a given mass loading. The calculated self-shielding factors have statistical uncertainties of up to 3% arising from limited computer run time.

It is concluded from Figure 1 that (1) PuO_2 particles within the range of 2,000 to 3,000 micron diameter yield an IPS bias commensurate with the waste form bias range determined in the 3,000 drum study, and (2) the self-shielding factor resulting from 2,000—3,000 micron particles does not significantly vary over the WRM weapons grade PuO_2 mass loading range of 0.25 to 16.0 grams. Hence, the active thermal-neutron interrogation-mode bias is principally due to individual particle size dimension and not large numbers of such particles.

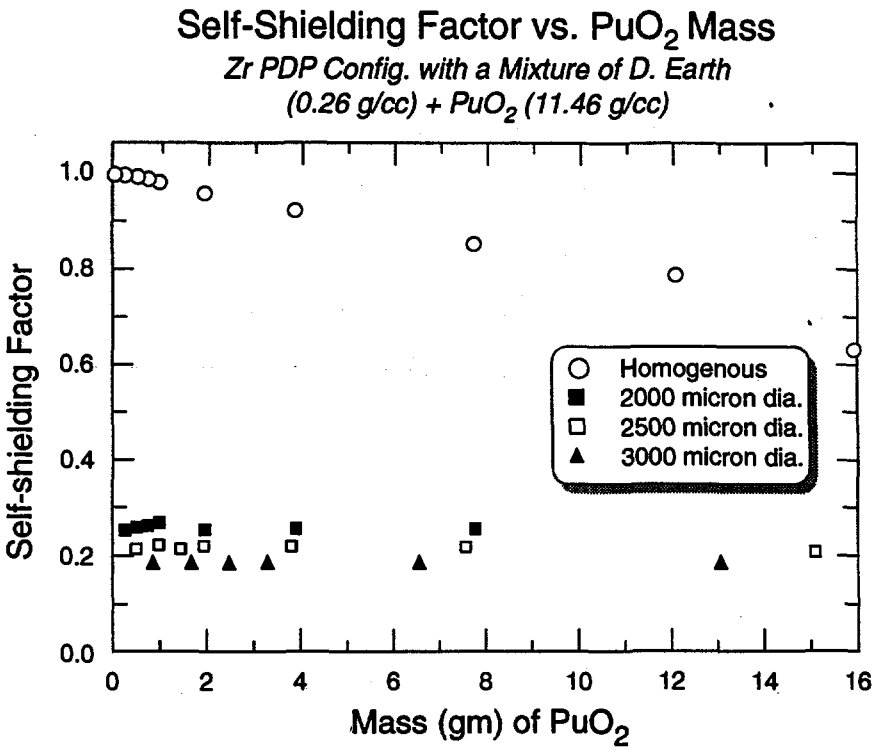

Figure 1. Plot of self-shielding factors verses PuO_2 mass.

To this point, discussion has centered on an IPS bias relative to neutron measurement modalities. It is constructive to evaluate the impact of the particle size determination with regard to gamma-based nondestructive assay techniques. To evaluate this effect, MCNP runs were performed to compute the number of gammas in a specific energy range leaving the surface of the WRM cylinder per gamma of the same energy originating within the PuO_2/diluent for both the homogeneous and heterogeneous WRM model cases. As an approximate indicator of this effect, we examine the ratio of 160 Kev gammas leaving the WRM surface per gamma originating in the homogeneous WRM model to that same parameter for the heterogeneous 2,000 micron diameter PuO_2 particle/diluent case. For a 0.24 gram PuO_2 WRM mass loading, which in the heterogeneous case is equivalent to five particles, this homogeneous/heterogeneous model ratio for 160 Kev gamma is 3.2. At a two gram WRM PuO_2 loading, approximately 40 particles for the heterogeneous configuration, this same ratio changes to approximately 3.3. For the 414 Kev gamma line at the 0.24 gram PuO_2 WRM mass loading, under these same conditions, the ratio is much less at 1.2. At two gram WRM PuO_2 loading, this same ratio is also about 1.2. In either case, increased PuO_2 particles sizes depress the yield of gammas relative to the homogeneous case, thus requiring that the gamma based NDA technique account for this effect.

DESIGN

The increased particle size (IPS) WRM specification required that large particles (granules) of PuO_2 with very accurately known mass be distributed within stainless steel cylindrical containers in a well known, uniform manner. The distribution is to be stable during normal transportation and usage. The Pu content of the IPS WRMs span a range from the sub-gram level to a few tens of grams. WRMs previously fabricated for the PDP were produced by blending finely divided PuO_2 powder in a diatomaceous earth matrix and encapsulating this mixture into cylindrical containers. This blended mixture was tested and found to be very uniform. However, IPS WRMs containing sub-grams to a few grams of Pu as millimeter-size PuO_2 granules would contain relatively few (20-50) granules, hence the established PuO_2-diatomaceous earth blending method might produce some WRMs in which the PuO_2 granules would not meet uniformity specifications. An alternate method for distributing the PuO_2 granules uniformly throughout the PDP cylinder volume was selected. This method uses a graphite felt material into which the PuO_2 granules are embedded. The graphite felt is purchased in 1/4 inch thick sheets and is readily cut into disks. The disks are drilled or punched to form holes into which PuO_2 granules are inserted. The impregnated disks are be stacked in the cylindrical container to yield a stable and uniform array of PuO_2 granules suspended in a low density matrix which does not appreciably interfere with either neutron- or gamma-based NDA measurements.

PuO_2 PARTICLE AND PROTOTYPE WRM FABRICATION

A. PuO_2 Preparation. PuO_2 is made in US facilities by either oxidizing Pu metal in air or by precipitating a Pu compound (oxalate or peroxide) from a solution and then calcining the compound in an air atmosphere. Both methods produce a fine PuO_2 powder with particles distributed through a range of typically 5 to 500 microns. The larger particles are usually quite fragile, breaking into smaller particles upon mechanical handling. We felt that large and mechanically tough PuO_2 particles could be made using methods employed to make uranium oxide or mixed Pu - U oxide (MOX) reactor fuel pellets. The fuel pellets are prepared by adding a small amount of organic binder to U oxide or MOX powder, pressing the powder in cylindrical molds at ~1,500 psi pressure, and then calcining the fuel pellets at 1600 ^{o}C for six hours. The resulting fuel pellets are extremely 'tough' and have a density above 90% of the theoretical density. The reactor fuel R&D group at the Los Alamos Plutonium Fabrication Facility (TA-55) had, as of October 1996, not made 'straight' PuO_2 fuel pellets, but agreed it would be feasible to do so and developed a successful method. After preparation, the PuO_2 fuel pellets (approximately 5 mm diameter and 10 mm long) were crushed by forcing through appropriately sized sieves, producing PuO_2 granules

meeting the size requirements determined via the MCNP modeling. Figure 2 is a photograph of the PuO_2 granules prior to final sizing. These granules have proven to be very 'tough' in that they can be stressed with tweezers or dropped with no evidence of fracturing. This toughness allows the granules to be inserted into holes in the graphite felt disks without concern of the granules breaking during fabrication or subsequent transportation or use.

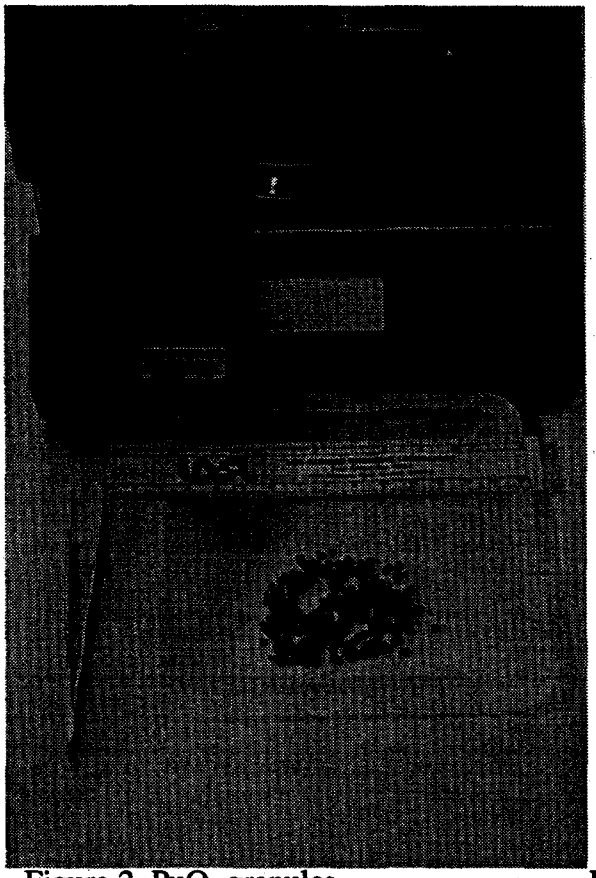

Figure 2. PuO_2 granules.

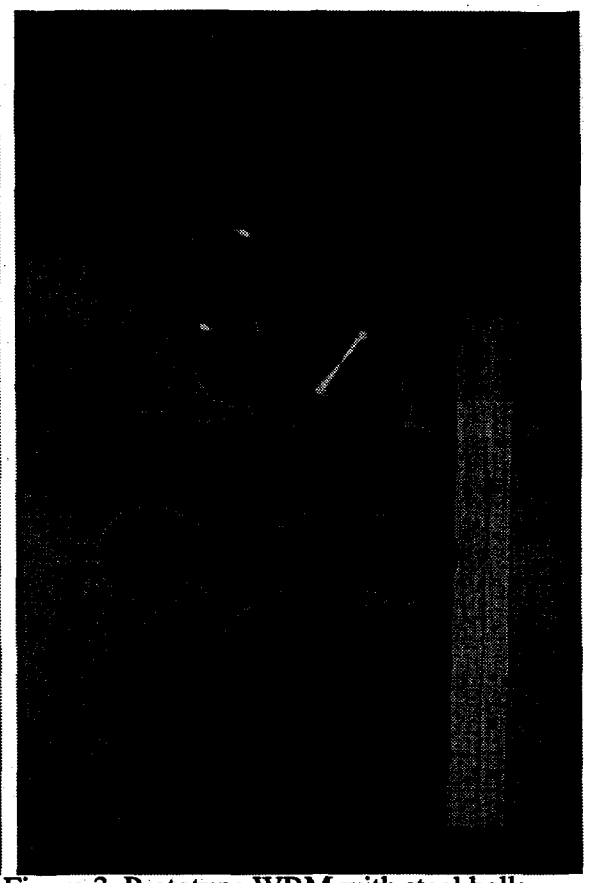

Figure 3. Prototype WRM with steel balls representing PuO_2 granules.

B. Prototype Fabrication. Of concern was whether or not the graphite disks would adequately retain the PuO_2 granules during the vibration and shock a WRM might experience during transportation and routine handling. To evaluate this stability, a prototype WRM was made using steel balls to represent PuO_2 granules. The steel balls allowed a prototype WRM to be fabricated 'on the bench' and transported and tested without the complications introduced by a WRM containing plutonium. Figure 3 is a photograph of the prototype constituents including the 1.75 inch O.D. steel cylinder plus its endcap, six graphite disks with embedded steel balls and 35

graphite disks used to space the six 'loaded' disks in a uniform vertical array. The six 'loaded' disks were prepared by drilling a ~0.17 inch hole through the disk and then pushing a 0.185 inch diameter steel ball into the hole. The graphite felt is a pliable, fibrous material that allowed the steel ball to be inserted quite gently. The felt fibers then fold in around the ball and hold it in place. The disks could be turned upside down without the steel ball falling out. To load the cylinder, five unloaded disks were introduced into the open end of the cylinder and pressed to the bottom with a large diameter rod. Then a loaded disk was introduced and pressed in place. This process was continued until the final 5 unloaded disks were introduced. Then the steel endcap was pressed in place and tack welded at four radial positions to complete the prototype assembly.

C. Prototype Testing. X-Ray imaging was used to evaluate the ability of the graphite array to withstand shock without unacceptable movement of the steel balls in the WRM configuration. An x-Ray image was photographed before subjecting the cylinder to shock. A segment of steel wire was affixed to the outside of the cylinder to provide a reference mark in the x-Ray image. Next the cylinder was dropped onto a hard floor from a height of 9 inches (the cylinders length) 15 times to simulate abusive treatment to a WRM. A second x-Ray image was photographed and then the cylinder was dropped 15 times from a 9 inch height onto it's side (same rotational orientation each drop). A third x-Ray image was photographed. Figure 4 shows the three x-Ray images. Note in the second photograph, the steel balls did move 'down' 5-10 mm in the tube after it was dropped. The third photograph shows two movements: first, the steel balls returned part way to their original axial position, and second, the steel balls moved slightly (~2 mm) towards the side of the tube receiving the impact upon dropping.

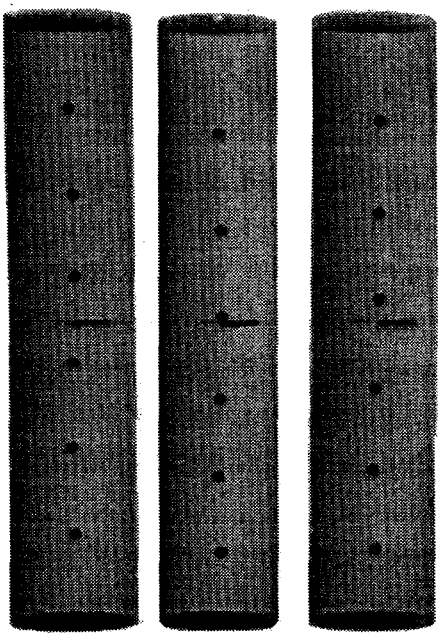

Figure 4. X-ray images of prototype WRM before and after drop testing.

It was apparent that the graphite disks were compressing upon the vertical dropping of the cylinder and that they were not decompressing after the drops were completed. Since the graphite disks were cut ~2 mm larger than the steel cylinder inside diameter, it appeared that friction provided by the somewhat oversized graphite disks prevented the disks from returning to their original position after each drop. A second prototype was prepared using graphite disks that were ~1 mm smaller in diameter than the cylinder inside diameter. This cylinder received the same testing as the first cylinder, and Figure 5 shows the results of this test. Note the second photograph shows a slight movement of the steel balls...averaging about 2 mm, and the third photograph shows a 'sideways' movement of the steel balls of 1 mm.

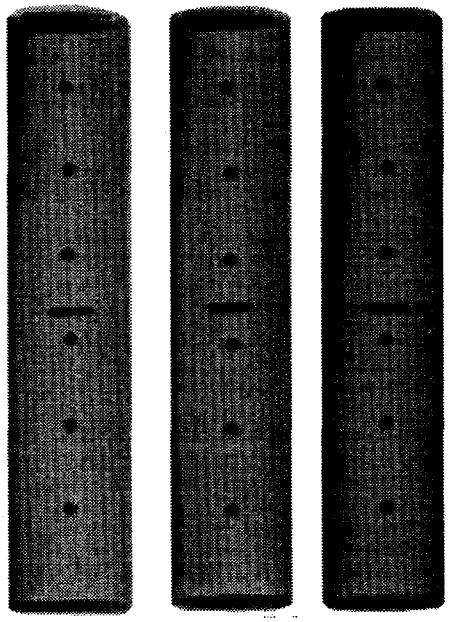

Figure 5. X-ray images of prototype WRM with loose fitting disks before and after drop testing.

The 1-2 mm movement is unlikely to introduce an observable bias to any known waste NDA measurement method used to determine the plutonium content of a WRM. Further, the steel balls had a mass approximating that of the PuO_2 granules on the most heavily loaded graphite disks, hence even less movement is expected in all but the most heavily loaded WRMs. The IPS WRMs will be fabricated using graphite disks (blanks and loaded with PuO_2) with diameter ~1 mm less than the cylinders.

Further stability tests will be conducted after the prototype WRM is subjected to vibrational stress simulating truck transportation of WRMs over a 2000 mile distance.

CONCLUSION

Through the techniques delineated, an increased PuO_2 particle size has been specified to yield and NDA system bias source reasonably approximating that of actual waste forms. Once specification of the particle size was complete, a means to produce PuO_2 particles within this range and subsequently secure them into a PDP WRM in a stable manner was devised. The completed WRM, configured in this manner, will be a valuable PDP program assessment tool for the evaluation of NDA system accounting and management of the self-shielding and self-attenuation bias source known to occur in waste forms

REFERENCES

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2 Performance Demonstration Program Plan for Nondestructive Assay for the TRU Waste Characterization Program, Revision 0, CAO-94-1045, March 1995, U.S. Department of Energy, Carlsbad Area Office.

3 MCNP computations performed by W. Yoon of Lockheed Martin Idaho Technologies Company, Idaho National Engineering Laboratory. PO Box 1625, Idaho Falls, Idaho 83515-2114