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USNRDL-TR-68-74

16 April 1968

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**RADIOACTIVITY RELEASE FROM RADIONUCLIDE
POWER SOURCES. VIIa. Dissolution Studies
of Plutonium Dioxide in the Ocean — 5- Months' Exposure**

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The work reported is part of a project sponsored by the Space Electric Power Office (SEPO), Atomic Energy Commission, under Contract Number AT(49-5)-2084.

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ABSTRACT

Post-ocean-exposure examinations have been made on plutonium dioxide microspheres recovered after a 5-month exposure on the bottom at a depth of 50 feet off San Clemente Island. These examinations established that approximately 70 percent of the microspheres became encrusted with marine growth.

Determination of the distribution of the microspheres and adsorbed activity in the ocean-bottom material was made in addition to post-exposure laboratory dissolution measurements, microscopic examination and X-ray diffraction measurements.

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SUMMARY

The Problem

A current USNRDL program involves the study of the dissolution behavior of SNAP fuel materials in seawater. Previous studies have been carried out under controlled laboratory conditions. In an effort to determine the effects of an in-situ ocean exposure, plutonium dioxide microspheres have been exposed on the bottom in a chamber at a depth of 50 feet off San Clemente Island. A post-ocean exposure examination has been carried out on microspheres recovered after 5 months.

The Findings

The most significant finding of this study is that a large fraction, approximately 70 percent, of the microspheres became encrusted with marine growth. Studies of the distribution of the microspheres and adsorbed activity in and on the bottom material indicated that both would remain localized. Measurement of the adsorbed plutonium activity indicated that very little of the released activity was adsorbed in comparison to the extent of adsorption in laboratory experiments.

Measurement of the post-ocean-exposure dissolution rates in the laboratory indicated that the dissolution rate of clean (unencrusted) microspheres was about the same as that of a laboratory control sample while that of the encrusted microspheres was about a factor-of-two less.

Microscopic examination of the microspheres showed no visible evidence of surface or structural changes. X-ray diffraction results indicated single phase stoichiometric $\text{PuO}_{2.00}$. Crush strength determinations were inconclusive because of the small number of microspheres used for the test.

I. INTRODUCTION

In the event that a radioisotope-fueled SNAP (Systems for Nuclear Auxiliary Power) Space unit, either intact or damaged, is placed accidentally in an ocean environment, potential release of the radioisotopic fuel material to the ocean may generate a radiological hazard. Studies of the nature and extent of such a release are required for assessment of the magnitude of the radiological hazard.

A current USNRDL program involves the study of the dissolution behavior of various SNAP radioisotopic fuel materials in seawater. In addition to controlled laboratory experiments, a test plan for in-situ ocean environment exposure of an uncontained fuel material, plutonium-238 dioxide microspheres, has been implemented.* In this test, an exposure chamber located on the ocean floor contains the plutonium dioxide fuel and ocean-bottom material; a water circulation system maintains the interior of the exposure chamber in equilibrium with continuously refreshed ocean-bottom water. The same circulation system provides pumping of effluent water from the exposure chamber to a shore station for radioactivity monitoring.

This report presents the results of a post-ocean-exposure examination of the plutonium dioxide microspheres after a 5 month exposure on the bottom at a depth of 50 feet off San Clemente Island.**

* L. W. Weisbecker and J. L. Mackin, "Basic Test Plan for Ocean Exposure Studies of SNAP Radioisotopic Fuel Material", U.S. Naval Radiological Defense Laboratory, USNRDL-LR-191, 7 July 1966.

** D. A. Kubose, "Basic Test Plan for Post-Ocean-Exposure Examination of SNAP Fuel Material", U.S. Naval Radiological Defense Laboratory, USNRDL-LR-67-86, 18 July 1967.

IIa. EXPERIMENTAL - DEPLOYMENT

Exposure Chamber and Pumping System

The conceptual design of the exposure chamber and pumping system is presented in the basic test plan (first footnote, page 1); a detailed description of the complete exposure system is contained in a private communication.* Figure 1 shows a sketch of the exposure chamber. The ID of the lower cylindrical section is 25.5 in. The overall height of the chamber is 31 in. The total volume of the chamber is approximately 100 liters.

Placement of Fuel in Exposure Chamber

The ocean-bottom material (six inch depth) placed in the lower section of the exposure chamber was taken from the pre-selected ocean-bottom location for the test. The plutonium dioxide microspheres (4.18 mg $\equiv$ 238 microspheres) were placed on the surface of the bottom material at the center of the exposure chamber. After placement of the microspheres, the top section of the exposure chamber was bolted on and the chamber lowered into the ocean.

Exposure Conditions

The exposure chamber was placed on the bottom at a depth of about 50 feet off the North Light Harbor area of San Clemente Island. The influx of fresh ocean-bottom water into the exposure chamber was such that there was a complete change of water volume every 1.7 min.

The average ambient water temperature was approximately 15°C.

Duration of Exposure

A total of four exposure chambers have been placed on the ocean-bottom. The first chamber was recovered after a period of approximately 5 months (140 days) and is the subject of this report. The other three chambers will be recovered at intervals of 5 months (i.e., the fourth chamber will have been in operation for 20 months when it is recovered).

* Private Communication, D. M. Elliott, "SNAP Fuel - Ocean Exposure Facility. Data Package and Operation Manual", Stanford Research Institute.

Fig. 1 Exposure Chamber

IIb. EXPERIMENTAL - RECOVERY AND ANALYSIS

Distribution of Fuel Material Within Bottom Material

Because of the possibility that the fuel distribution at the end of the test would be disturbed during transport of the recovered exposure chamber from San Clemente Island to NRDL, post-exposure sampling of bottom material in the exposure chamber was done at San Clemente Island. The samples, packaged in one-pint polyethylene frozen food containers and properly catalogued as to the relative positions from which they had been taken within the exposure chamber, were transported to NRDL for analysis.

Preliminary studies had indicated that individual plutonium dioxide microspheres can be isolated from large amounts of ocean-bottom material by monitoring of the Pu^{238} gamma rays. The gamma rays can be detected by a low-background scintillation counter even when a microsphere is in several hundred grams of bottom material. Therefore, each of the polyethylene containers was counted in the scintillation counter for determination of the presence of fuel material. Containers having count rates significantly above background were set aside for subsequent subdivision of their contents. After all the containers had been counted, the bottom material in each of a selected number of the containers having the highest count rates was subjected to successive subdivision for isolation of the microspheres. During the final stages of subdivision a combination of microscopic examination and radioactivity detection by a Geiger-tube probe was used for location of individual microspheres.

After recovery of the microspheres the bottom material was examined for the amount of adsorbed plutonium activity.

Activity Adsorbed on Bottom Material

Previous studies¹ have indicated that a large fraction of the radioactivity released from the microspheres will be adsorbed on ocean-bottom material. In the present studies sections of ocean-bottom material which contained large, intermediate, and small numbers of microspheres were examined for adsorbed plutonium activity. Sections which contained no microspheres were also examined.

Because of the extremely low specific activity of adsorbed plutonium expected, a radiochemical analysis was performed on the samples of bottom material to isolate the plutonium activity for alpha counting. The following radiochemical procedure, a modification of that of de Bortoli,² was used.

The plutonium activity was extracted from 500 g of bottom material with hot concentrated hydrochloric acid for several hours. After cooling the solution was filtered through a #42 Whatman filter paper, the residue washed twice with 50 ml of 1N hydrochloric acid and the wash solution filtered through a #42 Whatman filter paper. All the filtrates were combined and evaporated to a volume of 50 ml. The precipitate resulting from addition of ammonium hydroxide to the 50 ml volume was separated by centrifugation and was redissolved and reprecipitated twice with ammonium hydroxide. This final precipitate was dissolved in 50 ml of concentrated nitric acid and the resulting solution evaporated to dryness. The dry residue was dissolved in 20 ml of distilled water containing a few drops of concentrated nitric acid and the resulting solution diluted to 25 ml in a volumetric flask. A 1 ml aliquot of this solution was counted in a liquid scintillation counter by a previously described method.³

Post-Exposure Laboratory Solubility Test

A fraction of the recovered microspheres was immersed in 1 liter of seawater under laboratory conditions for determination of their post-exposure dissolution rate. This rate was compared to that of a laboratory control sample from the same production batch. The experimental techniques used have been reported elsewhere.^{4,5}

Other Tests

Several of the recovered microspheres were sent to Mound Laboratory, Miamisburg, Ohio, for photomicrography, crush strength tests and X-ray diffraction studies to check fuel stoichiometry.

III. RESULTS AND DISCUSSION

Recovery of Microspheres from Ocean-Bottom Material

Figure 2 shows how the bottom material within the exposure chamber was sectioned (layer 1 represents the topmost layer). Each layer was about 1.5 in. thick and 25.5 in. in diameter. The total volume of bottom-material was approximately 50 liters.

Fig. 2 Distribution of Microspheres Within Ocean-Bottom Material

TABLE 1

Distribution of Recovered Microspheres Within Exposure Chamber

Section *	Number of Clean Microspheres	Number of Encrusted Microspheres
1Ca	0	24
1Db	1	0
1Eb	0	2
1Gb	0	1
1Hb	2	2
1 Center	1	1
2Aa	0	2
2Ba	1	12
2Ga	1	1
2Ha	1	1
3Aa	4	4
3Ba	3	1
3Ca	3	0
3Da	1	0
3Ea	1	0
3Fa	1	0
3Ga	1	1
3Ha	1	4
3Db	0	1
3Gb	0	1
4Aa	2	0
4Ba	2	0
4Ea	0	1
4Ga	1	0

* Refer to Figure 2. a represents inner ring; b represents outer ring.

The numbers shown in the sections of each figure designate the number of microspheres found in each section.* Comparison of the layers in Figure 2 shows that the majority of the microspheres remained near the surface of the bottom material. Only 6 microspheres migrated down as far as about 5 inches from the bottom-material/seawater interface.

86 of the 238 implanted microspheres were isolated from the bottom material. The sections processed to recover microspheres were selected from sections which showed significant count rates (see Section IIb). Of these 86 microspheres 59 were covered (encrusted) with marine organisms. Table 1 shows the distribution of the encrusted and clean (unencrusted) microspheres. Table 2 shows the distribution and the ratio of the number of encrusted to clean microspheres in each layer. The fact that the ratio of encrusted to clean microspheres is much greater near the bottom-material/seawater interface than further below suggests that encrustation of the microspheres at least partially might be responsible for the distribution observed in Fig. 2. That is, once a microsphere becomes encrusted its lower effective density slows or prevents its migration downward.

TABLE 2

Distribution and Ratio of Number of Encrusted
to Clean Microspheres in Each Layer

Layer	Number of Clean Microspheres	Number of Encrusted Microspheres	Ratio (Encrusted/ Clean)
1	4	30	7.5
2	3	16	5.3
3	15	11	0.7
4	5	2	0.4

* The sections listed in Table 1 were the only ones subdivided for recovery of microspheres. The numbers of microspheres in the remaining sections of Fig. 2 were approximated from their respective count rates (see Section IIb) by means of the correlation of the count rates of the sections listed in Table 1 with the total number of microspheres actually found in them. This resulted in accounting for 230 of the 238 microspheres implanted.

Figure 3 shows photomicrographs, taken at NRDL, of some of the encrusted microspheres. Microscopic examination of several encrusted microspheres was made by Dr. Burton E. Vaughan of this Laboratory. The following are his comments.*

"Wet mount preparations were made by placing a drop of seawater containing one Pu^{238} aggregate [encrusted microsphere] in the concave area of a hollow ground slide. By applying slight pressure to the overlaying coverslip the aggregate broke apart. Preparations were viewed microscopically at 100X, 430X and 970X.

The microsphere was clearly visible in each aggregate and it appeared to be little altered by submergence for 5 months in the sea. Numerous small diatoms were adherent to the uniformly smooth surface of the Pu^{238} sphere. The aggregates were composed of sand grains (quartz or silica) and organic detritus, which was lightly cemented around the microsphere by a dense matrix of mucilage and small diatoms. Areas of the broken aggregate most densely populated by the diatoms appeared yellow-brown in color, due most likely to a characteristic organic pigment manufactured by these cells.

The predominant organism present was tentatively identified as Nilzschia sps. This is a relatively small cell measuring 2-10 μ and it is motile when not existing in colonial or epiphytic form. It is widely distributed, occurring most frequently as a dense mat. The cells were connected in various patterns by means of a mucilaginous secretion.

Appearing in the layer aggregates were several centric diatoms, measuring 24-40 μ . These were circular in form, with radial markings. A few elongate, pennate cells were also observed and identified as Navicula sps. Occurring in one preparation were 2 clusters of elongate, tapering, tube structures, yellow-brown in color, estimated to be 200 μ in length. These were of biological origin, and probably represented a colonial coelenterate or a bryozoan.

* Private communication, Dr. B. E. Vaughan, Sept. 1967.

Bacteria were present in all preparations, and in at least two of the sedimentary aggregates, small unicells were present, 2-5 μ in size. The unicells were definitely yellow-green in color and showed active mobility by means of a single flagellum. These latter forms may be the unicellular chlorophyte, Nannochloris sps., but positive identification could not be made at this time.

There seems to be little question that the Pu^{238} particle had been surrounded by an organic layer, which seems to have cemented sand grains to its surface. However, it is not clear from these preliminary observations that most of the organisms found are typically found in sediment. Centric diatoms for example are usually found in upper water layers.

An examination of the photomicrograph showing a gastropod shell (Fig. 3J) with a similar aggregate attached to it indicated this was adventitious: The shell had many small holes bored through it, clearly showing that its host had long since died."

Adsorbed Activity

The bottom material in various representative sections (some from Table 1, and some in which no microspheres were found) was assayed for adsorbed plutonium activity after removal of all microspheres from these sections. Table 3 shows the amount of adsorbed activity found in these sections. The number of microspheres contained in these sections is also shown. There appears to be a definite correlation between the number of microspheres present in a section and the amount of adsorbed activity in that section (constancy of entries in last column, Table 3). However, as shown below, the post-exposure dissolution rate for the encrusted microspheres is about one half that of the clean microspheres. Since most of the microspheres in the upper layers are encrusted (see Table 2), it might be expected that the adsorbed activity per bead in the bottom material for the upper layers should be about half that for the lower layers. The fact that it is not can be accounted for if one accepts the hypothesis that part of the adsorbed activity in the top layer comes from the clean microspheres located in the top layers before they migrate to the lower layers or from the encrusted microspheres before they become encrusted. The latter appears to be the more likely explanation since the number of clean microspheres in the lower layers is small.

100 μ

Fig. 3 Photomicrographs of Encrusted Microspheres

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Fig. 3 Continued

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Fig. 3 Continued

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TABLE 3

Plutonium Activity Adsorbed on Bottom Material

Section	Number of Microspheres	Adsorbed Activity		Weight % of Microspheres Adsorbed
		Pu ²³⁸ (c/m)	PuO ₂ Equivalent (μg)	
1Ca	24	2185	1.0×10^{-4}	2.4×10^{-5}
1Eb	2	125	5.7×10^{-6}	1.7×10^{-5}
2Ba	13	950	4.3×10^{-5}	1.9×10^{-5}
3Ba	4	248	1.1×10^{-5}	1.6×10^{-5}
4Aa	2	150	6.8×10^{-6}	1.9×10^{-5}
3Cb	0	< 100	$< 5 \times 10^{-6}$	-
4Ca	0	< 100	$< 5 \times 10^{-6}$	-

Post-Exposure Laboratory Solubility

Figure 4 presents the total release of plutonium dioxide, expressed as μg released per mg of plutonium-dioxide microspheres, from the laboratory control sample and from the recovered clean and encrusted microspheres.

The weights of the clean and encrusted microsphere samples were calculated from the respective number of microspheres in each sample assuming that the number-size distributions of the microspheres in these samples were the same as that of the microspheres in the original sample of 238 microspheres. A value of 10.1 g/cm³ was used for the density of the microspheres in relating size to weight.

Table 4 shows the release rates, in μg/mg/day, obtained from the slopes of the total release curves in Fig. 4. The release rate for the clean microspheres is about the same as that of the control sample while the rate for the encrusted microspheres is about a factor-of-two less. The lower release rate for the encrusted microspheres is probably due to the diffusion barrier of marine growth around the microspheres.

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Fig. 4 Total Release of PuO_2 from Laboratory Control, Clean and Encrusted Microspheres

TABLE 4

Dissolution Rate of Clean and Encrusted Microspheres

Sample	Dissolution Rate - 10^{-3} $\mu\text{g}/\text{mg}/\text{day}$							
	0-20d*	20-60d	60-100d	100-140d	140-160d	160-200d	200-240d	240-280d
Control	1.33	1.35	1.35	1.48	1.60	1.60	1.60	1.60
Clean	———— in ocean —————				1.20	1.30	1.33	1.43
Encrusted	———— in ocean —————				0.48	0.61	0.65	0.76

*
d = days

It is of interest to examine the relation between the amount of plutonium activity adsorbed on bottom material and the net amount released to seawater from the microspheres. Previous work¹ has indicated that in a static system (i.e., microspheres placed on bottom-material contained in a beaker of seawater with no agitation) essentially all of the released plutonium activity is adsorbed on the bottom material. In contrast, based on the release rate of encrusted microspheres (Table 4) and the amount of activity adsorbed (Table 3, assuming a linear relationship between the number of microspheres and amount of activity adsorbed), only a fraction of a per cent of the activity released, assuming unchanged total dissolution rate, was adsorbed by the bottom material. This discrepancy can be accounted for at first glance by the observation that in the present mode of exposure the seawater is continuously being refreshed at a relatively rapid rate (1.7 min. turn-over time). The rate of adsorption is apparently much lower than the rate at which the released activity is swept away by the flow of fresh seawater.

The above explanation is not entirely satisfactory. For example it is not known exactly how deep the majority of the microspheres were buried. If they were buried to any extent, one would expect that more of the released activity would have been adsorbed on the bottom material than was measured, even with limited circulation of water in the upper layer. It would also be of interest to know the isoadsorption lines around a buried microsphere and the saturation limit of adsorbed activity on the bottom material.

It is evident that several questions remain to be answered. Analysis of the contents of the next exposure chamber, in addition to related laboratory experiments, will be directed to answering these questions.

Other Tests

Two sets of samples, one consisting of 5 clean microspheres and the other consisting of 10 encrusted microspheres (so identified), were sent to Mound Laboratory, Miamisburg, Ohio for further tests. The following is the correspondence received from them.*

"Two samples of material which had been exposed to natural ocean environment were received from USNRDL. One sample, which appeared to be free of any ocean deposits, consisted of four intact and one crushed microsphere. The other sample contained only two recognizable microspheres, which had some apparent ocean deposit on their surfaces, accompanied by what appeared to be "debris" from the ocean bottom. All of the particles from both samples were examined microscopically and photomicrographs were taken. The microscopic examination did not reveal any apparent surface or structural changes in the microspheres except for the ocean deposits on two of the six identifiable microspheres.

X-ray diffraction results from two of the "clean" microspheres were indicative of single phase stoichiometric $\text{PuO}_{2.00}$. The X-ray data however, cannot exclude the possibility of the presence of some phase in the form of a physical admixture of the order of 5 percent or less.

Because of the inherent difficulties in handling microspheres only three of the microspheres remained to be transferred for crush strength analysis. Two of these microspheres were those which had ocean deposits on their surfaces. The results from the crush strength analysis however were inconclusive because it was impossible to determine whether or not the particles had been crushed.

* Private communication from Mound Laboratory; Inter-Office correspondence from Gary N. Huffman to Dr. Richard E. Vallee, 8 Feb 1968.

It was decided to subject the material which appeared to be ocean bottom "debris" to a concentrated hydrochloric acid leach for twenty-four hours in order to ascertain whether any PuO_2 microspheres might be obscured in this portion of the sample. Eight $^{238}\text{PuO}_2$ microspheres were recovered from this portion of the sample completely free of any surface deposits after this acid treatment. A visual inspection of the leached microspheres showed no noticeable surface or structural changes. A white solid residue which was not identified accompanied the microspheres. An attempt was made to separate the free microspheres from this residue by density difference using a liquid flotation method. Only one microsphere was successfully recovered and subjected to crush strength analysis. A crush strength of 585 grams was obtained for this microsphere. This is in comparison to an average value of 295 grams for current production grade $^{238}\text{PuO}_2$ microspheres. No conclusions should be drawn from this single value of 585 grams. In order to obtain more meaningful data, further samples should be studied."

IV. CONCLUSIONS

The most significant finding of this study is that a large fraction of the plutonium dioxide microspheres became encrusted with marine growth. The occurrence of this phenomenon is beneficial in the sense that it probably lowers the rate of dissolution of plutonium activity from the microspheres; on the other hand it raises the question of whether or not it would increase the probability of a microsphere's entering the marine food chain.

The observed distribution of microspheres within the bottom material indicates that the microspheres do not migrate to a significant extent. This observation, in addition to the finding that the adsorbed activity distribution is the same as that of the microsphere distribution, indicates that in the event plutonium dioxide microspheres are accidentally placed on the ocean bottom the microspheres and adsorbed activity would remain localized. This localization could of course be modified by strong bottom current agitation and scouring of the bottom material.

V. FUTURE PLANS

Recovery and analysis of the three remaining exposure chambers will provide further information such as changes in the microsphere distribution, changes in extent of encrustation of the microsphere and changes in post-exposure solubility rates. Emphasis will be placed on providing more detailed information regarding microsphere and adsorbed activity distributions. Related laboratory experiments will be conducted to provide information on items such as isoadsorption lines and saturation limit of adsorbed activity on bottom material.

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POWER SOURCES. VIIa. Dissolution Studies of
Plutonium Dioxide in the Ocean--5-Months' Ex-
posure, by D. A. Kubose, M. G. Lai, H. A. Goya,
and H. I. Cordova 16 April 1968 32 p.
tables illus. 5 refs. UNCLASSIFIED

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depth of 50 feet off San Clemente Island.
These examinations established
that approximately 70 percent
of the microspheres became
(over)

1. Seawater
2. Solubility
3. Plutonium dioxide
4. Reactor fuels

- I. Kubose, D. A.
- II. Lai, M. G.
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Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) U. S. Naval Radiological Defense Laboratory San Francisco, California 94135		2a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED	
		2b. GROUP	
3. REPORT TITLE RADIOACTIVITY RELEASE FROM RADIONUCLIDE POWER SOURCES. VIIa. Dissolution Studies of Plutonium Dioxide in the Ocean--5-Months' Exposure			
4. DESCRIPTIVE NOTES (Type of report and inclusive dates)			
5. AUTHOR(S) (First name, middle initial, last name) Donald A. Kubose Herman I. Cordova Ming-Gon Lai Harry A. Goya			
6. REPORT DATE (Distribution) 29 August 1968		7a. TOTAL NO. OF PAGES 32	7b. NO. OF REFS 5
8a. CONTRACT OR GRANT NO.		9a. ORIGINATOR'S REPORT NUMBER(S) USNRDL-TR-68-74	
b. PROJECT NO. SEPO, AEC, AT (49-5)-2084			
c.		9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)	
d.			
10. DISTRIBUTION STATEMENT This document has been approved for public release and sale; its distribution is unlimited.			
11. SUPPLEMENTARY NOTES		12. SPONSORING MILITARY ACTIVITY Atomic Energy Commission Washington, D. C. 20545	
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DD FORM 1473

1 NOV 65

(PAGE 1)

S/N 0101-807-6801

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14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Plutonium dioxide Solubility Ocean SNAP fuel						

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