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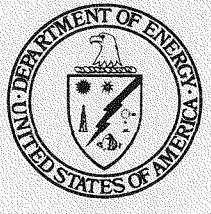
$^{238}PuO_2$ Fines Generation in Radioisotopic Heat Sources

Paul E. Teaney

June 23, 1983



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Contents

Abstract

Fuel aging studies were performed on the fuel form (plutonium-238 dioxide and yttrium) used in the Milliwatt Generator Radioisotopic Heat Source to determine the possibility of fuel degradation and of the resultant generation of respirable fines. In addition to long-term thermal aging of the fuel, evaluations included the effects of thermal ramping of the aged fuel to 1000°C and of impacting thermally hot (450°C) heat sources at 150 m/sec after thermal aging.

Introduction

Radioisotopic heat sources and radioisotopic thermoelectric generators (RTG's) are used in applications where reliable, long-term, localized heating or low-wattage power is required and access is difficult. These devices utilize the heat generated by decay of a radioisotope either directly for heating or indirectly by converting the heat to electrical energy through the use of thermoelectric material.

The most widely used radioisotopic fuel in such units is plutonium-238 dioxide. The plutonium-238 isotope is primarily an alpha emitter; thus, extensive radiological shielding is unnecessary. Its 87.79-yr half-life makes it ideal for long-term, high-power sources. The dioxide form of this isotope has good thermal stability and high specific power. However, since plutonium-238 is biologically hazardous if inhaled, the respirable fines content of the fuel must be kept as low as possible.

Long-term compatibility between the plutonium-238 dioxide and the tantalum alloy used to contain the fuel is enhanced by adding yttrium to the fuel when the heat source is fabricated [1]. This fuel form ($^{238}PuO_2$ and yttrium) has been used in several applications, such as the Radioisotopic Thermal Energy (RITE), the Radioisotopic Heater Unit (RHU), and the NAVY and Milliwatt Generator (MWG) heat sources.

This $^{238}PuO_2$ fines generation study was conducted at Mound as a part of a series of compatibility, pressure burst, and impact studies conducted for Sandia National Laboratories, Albuquerque (SNLA) to ensure the safety of the MWG heat source. These data may also apply to the other heat sources similarly fueled.

The fuel used in this application was hydroxide-precipitated PuO_2 shards, from 53 to 500 μm in diameter. The shards received their final sintering at 1600°C for 2 to 4 hr (depending on quantities present), during which time they also underwent oxygen-16 exchange. Yttrium was added to the fuel in a quantity to reduce the stoichiometry of the PuO_2 to $PuO_{1.75}$ during a 1-hr heat treatment at 1350°C.

The fuel aging studies reported here were performed to determine the possible degradation of the fuel form used in the MWG Radioisotopic Heat Source under both normal and accidental conditions and the resultant generation of respirable

fines.* These data are necessary in the unlikely event that the heat source is accidentally breached. In addition to long-term thermal aging of the fuel, evaluations included determining the effects on the PuO_2 particle size of thermal ramping of the aged fuel to 1000°C and of impacting a thermally hot (450°C) heat source at 150 m/sec after thermal aging.

The matrix for this study is shown in Table 1. The study, originally designed for fines determination in the <5 μm and the 5 to 10 μm size ranges, was later modified to include the 10 to 20 μm particle size range. As the tests progressed, proposed aging times for the specimens were extended to obtain longer-term data. Data on plutonia fines generation were obtained for specimens aged at 450°C for up to 27 mo after a 1-hr at 1350°C pretreatment.

Results

Metallography of the fuel was performed and the microstructures of the fuel at each stage of aging were compared. (Because the pressure mounting technique used for metallographic processing of the samples caused some shard breakup, the photomicrographs should not be used to estimate the generation of fines. Fines determinations were made using a demonstrated analytical technique [3].) Comparison of the fuel microstructure prior to testing (Figure 1) with the microstructure of the fuel after pretreatment (Figure 2) showed that, during pretreatment, helium in the fuel migrated to the grain boundaries, forming small round bubbles which delineated the boundaries. The two-phase appearance of several of the fuel shards in both the aged and pretreated specimens is indicative of substoichiometric fuel. Previous analyses have indicated that the lighter (acicular-structured) phase is PuO_2 and the darker phase is Pu_2O_3. Examples of the substoichiometric fuel are particularly evident in the etched photomicrographs in Figure 2.

Figure 3 shows fuel aged for 6 mo at 450°C. The delineation of the grain boundaries by helium bubbles appeared more pronounced than immediately after pretreatment. However, the microstructure did not indicate fines generation during the first 6 mo of aging. If breakup occurred along the delineated grain boundaries, the estimated grain size of the fuel would only account for increases between 10 and 90 μm. No additional changes in microstructure were noted in the fuel after 9, 15, and 27 mo, although transgranular fracture of the fuel shards was evident after 27 mo of aging and impact at 150 m/sec, 450°C. In the thermally ramped (to 1000°C) sample, the fuel microstructure appeared no different than aged fuel (Figure 3).

Plutonia fines data for the studies are summarized in Table 2. Comparison of data for the baseline fuel with the pretreated fuel indicated that no fines were generated during pretreatment. The baseline fuel was rechecked after aging

*Respirable fines are those particles with an equivalent unit density aerodynamic diameter of less than 10 μm, which for PuO_2 with a particle density of 11.46 g/cm^3 translates to a PuO_2 particle diameter of 3 μm. For the Mariner-Jupiter-Saturn RHU, respirable fines were considered to be all particles less than 4.5 μm in diameter [2].

Table 1 - FUEL AGING STUDIES

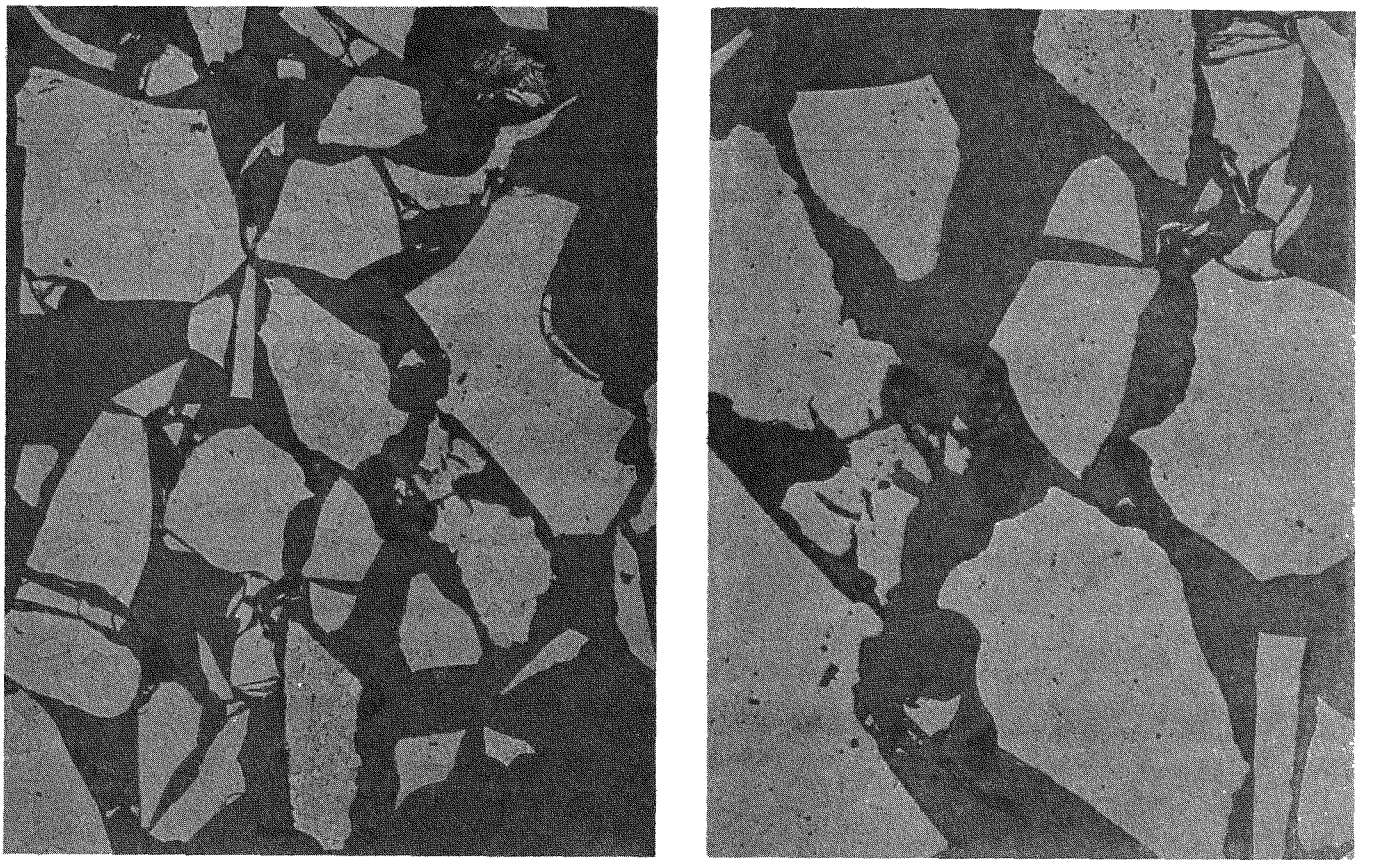
Sample Description	Type of Sample[a]	No. of Samples	Aging Conditions	Analyses Performed
Baseline Fuel	A	2	-	<5μ fines, fuel microstructure
Pretreatment Effects	B	2	-	<5μ fines, fuel microstructure
Impact Effects on Unaged Fuel	C	2[b]	-	<5μ fines, >5μ fines distribution
Aging Effects	C	2	6 mo at 450°C	<5μ fines, fuel microstructure
	C	2	≥ 9 mo at 450°C	<5μ fines, fuel microstructure
	C	2	≥15 mo at 450°C	<5μ fines, fuel microstructure
	C	1	≥27 mo at 450°C	<5μ fines, fuel microstructure
Impact Effects on Aged Fuel	C	2	≥27 mo at 450°C	<5μ fines, >5μ fines distribution
Thermal Ramp (to 1000°C) Effects on Aged Fuel	C	2	≥27 mo at 450°C	<5μ fines, fuel microstructure

[a] A - PuO_2 shards, no Y.

B - PuO_2 shards with Y, encapsulated in T-111 alloy liner and strength member, pretreated (1 hr at 1350°C).

C - PuO_2 shards with Y, encapsulated through clad, pretreated in strength member.

[b] Impact data agreed with previous impact data; therefore, second capsule was used for other tests.



125X 250X

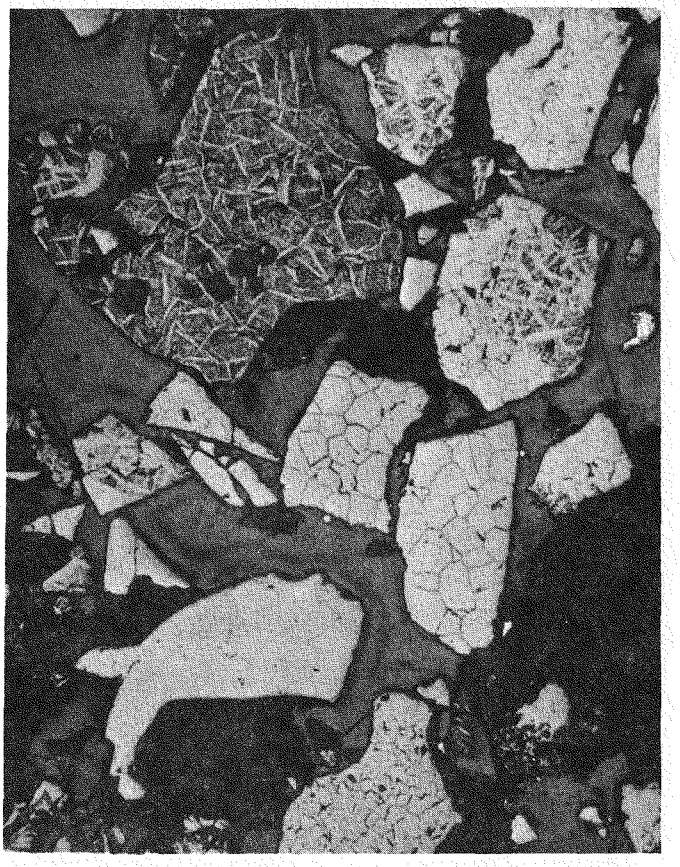
FIGURE 1 - Fuel microstructure prior to testing (single phase with no helium bubbles).

10 mo in a 20 to 30-g batch and no significant increase in fines was observed. Comparison of data for an impacted specimen (MF-29C) with the baseline fuel (P-438) indicated an increase in <5 μm fines from 3.83×10^{-3} to 4.08×10^{-2} wt %. However, there was no increase in the 5 to 10 μm particle size range.

Comparison of data for the baseline fuel with pretreated fuel aged for 6, 9, 15, and 27 mo at 450°C indicated a slight initial generation of fines during the first 6 mo of aging. No additional fines were generated through 27 mo of aging at 450°C in the <5 μm and 5 to 10 μm size ranges. However, fines in the 10 to 20 μm size range increased approximately one order of magnitude between 15 and 27 mo of aging at 450°C.

MF-34C and MF-36C were thermal ramped to 1000°C (for ∿5 min) after aging for 27 mo at 450°C. There was no increase in the number of <5 μm or 10 to 20 μm fines as a result of the thermal ramp. The fines present in the <5 μm size range occurred during the first 6 mo of aging at 450°C and the fines present in the 10 to 20 μm size range apparently occurred during the 15 to 27 mo of thermal aging. In the 5 to 10 μm size range, one specimen (MF-34C) showed an increase while the other (MF-36C) did not. The results for this size range are, therefore, inconclusive.

Two specimens (MF-30C and MF-31C) were aged for 27 mo at 450°C and then impacted at 450°C at 150 m/sec. No additional <5 μm fines resulted from impacting the aged fuel. However, there was a slight increase in 5 to 10 μm fines and a



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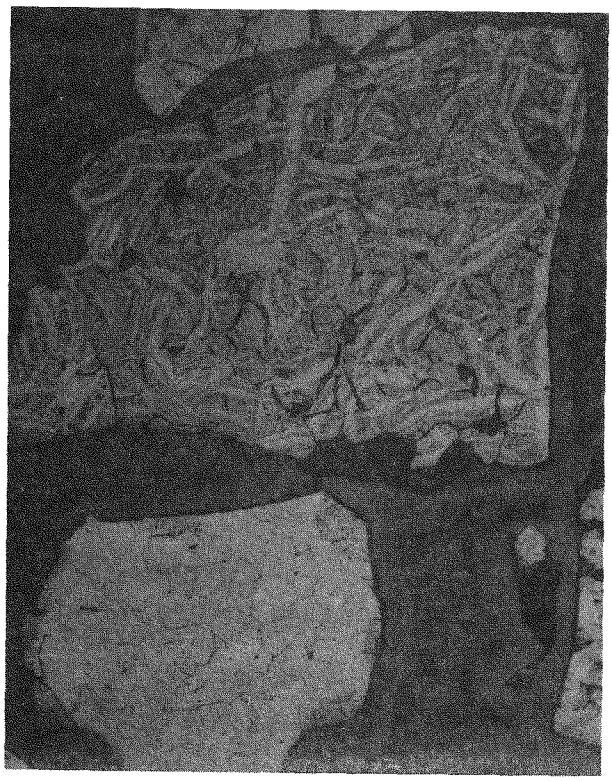
210X (Etched)

FIGURE 2 - Fuel microstructure after pretreatment
(two phases with helium migrated to grain boundaries).



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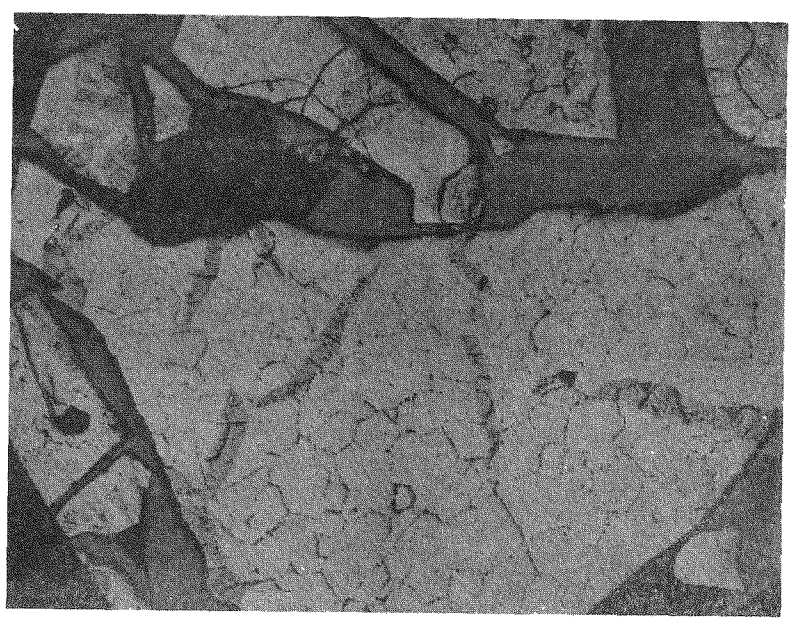

220X

FIGURE 3 - Fuel microstructure after aging for six months.

Table 2 - PLUTONIA FINES DATA FROM FUEL AGING STUDIES

Sample Number	Test Conditions	Wt % ^{238}Pu Fines		
		<5 μm	5-10 μm	10-20 μm
P-438	Baseline Fuel for MF-29C through -38C	3.83×10^{-3}	4.3×10^{-4}	a
P-445	Baseline Fuel for MF-39C through -47C	1.73×10^{-3}	5.9×10^{-4}	a
MF-37C	Pretreated Fuel	6.5×10^{-4}	4.9×10^{-4}	a
MF-38C	Pretreated Fuel	1.36×10^{-3}	7.0×10^{-4}	a
MF-29C	Impacted Fuel (149 m/sec at 450°C)	4.08×10^{-2}	1.0×10^{-4}	a
P-445 (aged)	Aged 10 mo in a 20-30 g batch	4.96×10^{-3}	6.6×10^{-4}	a
MF-39C	Aged 6 mo at 450°C (after pretreatment)	1.53×10^{-2}	2.98×10^{-2}	1.32×10^{-2}
MF-40C	Aged 6 mo at 450°C (after pretreatment)	2.25×10^{-2}	1.74×10^{-2}	1.24×10^{-2}
MF-41C	Aged 9 mo at 450°C (after pretreatment)	1.60×10^{-2}	3.34×10^{-2}	4.34×10^{-2}
MF-42C	Aged 9 mo at 450°C (after pretreatment)	2.52×10^{-2}	2.08×10^{-2}	3.14×10^{-2}
MF-43C	Aged 15 mo at 450°C (after pretreatment)	2.27×10^{-2}	1.01×10^{-2}	1.22×10^{-2}
MF-44C	Aged 15 mo at 450°C (after pretreatment)	9.33×10^{-3}	1.13×10^{-2}	3.11×10^{-2}
MF-34C	Aged 27 mo at 450°C and thermal ramp to 1000°C (after pretreatment)	2.25×10^{-2}	1.91×10^{-1}	2.37×10^{-1}
MF-36C	Aged 27 mo at 450°C and thermal ramp to 1000°C (after pretreatment)	1.56×10^{-2}	1.94×10^{-2}	1.84×10^{-1}
MF-30C	Aged 27 mo at 450°C and impacted 150 m/sec at 450°C (after pretreatment)	3.10×10^{-2}	5.50×10^{-1}	1.35×10^{1}
MF-31C	Aged 27 mo at 450°C and impacted 150 m/sec at 450°C (after pretreatment)	1.10×10^{-2}	1.86×10^{-1}	1.40×10^{1}
MF-33C	Aged 27 mo at 450°C (after pretreatment)	7.47×10^{-3}	6.86×10^{-3}	1.08×10^{-1}

[a]Determinations in this size range were not performed during the initial phase of this study.

significant increase (from ∿0.1 to between 13.5 and 14.0 wt %) in particles in the 10 to 20 μm size range. The increase appears to be normal for impacted fuel and thus appears to be independent of thermal aging. (Unfortunately, determinations in this size range were not being made when the baseline fuel and initial impacted specimen were analyzed.) Similar results were later obtained for heat sources containing unaged fuel. Four heat source specimens were impacted at ∿150 m/sec at 450°C shortly after fabrication using unaged fuel. Results for the 10 to 20 μm particle size distribution ranged from ∿2 to 30 wt %. Results for the aged specimens after impact fall approximately midway in this range.

Summary

No <5 μm or 5 to 10 μm fines were generated during the 1 hr at 1350°C pretreatment prior to thermal aging. There was an increase in <5 μm and 5 to 10 μm fines during the first 6 mo of thermal aging and an increase in <5 μm fines during the impact of specimens containing unaged fuel. No further increases in the size range containing respirable fines (<5 μm) were observed through 27 mo of thermal aging or thermal ramping or impacting of aged fuel specimens. The thermal ramp of the aged fuel to 1000°C apparently does not generate fines; however, results for the 5 to 10 μm size range are inconclusive. Impacted aged specimens showed some increase in 5 to 10 μm fines. Also, there was a considerable increase in 10 to 20 μm particles. However, more recent data indicated that this is characteristic of impacted specimens and that thermal aging most likely did not contribute to the increase.

Acknowledgement

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