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Gas Generation Measurements Of Scrap Pu/U Materials Using A Bell Jar

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ABSTRACT

A bell jar is used to determine containment vessel pressurization due to gas generation from plutonium/uranium materials. Seventy eight food pack cans containing plutonium and uranium oxide bearing materials have been tested to date. Minimal change in pressure (increase or decrease) occurred in fifty one cases, depressurization occurred in seventeen cases, and pressurization occurred in ten cases. Pressurization is considered to be linked to the presence of certain impurities such as magnesium oxide.

I. INTRODUCTION

The primary purpose of the bell jar is to collect and measure gas generation from a container (e.g., food can) of plutonium bearing materials in support of understanding shipping container pressurization. The bell jars have been designed and sized to accommodate most existing storage containers and collect out gassing without repackaging. The pressurization of the bell jar can be readily extrapolated to the conditions inside the shipping container containment vessel during a shipment. Bell jar testing is used in conjunction with current plutonium surveillance (e.g. lid deflection measuring device) to demonstrate that the containers bearing plutonium material may be shipped safely. Statistical sampling of similar materials can facilitate the release of larger inventories for shipment.

The 'bell jars' are heavy-duty multi-use, leak tight steel vessels in which a container of plutonium may be tested for out gassing. The bell jar is instrumented to measure pressure and temperature to very tight tolerances. Temperature and pressure data are continuously recorded through a data acquisition system to a lap top computer. The threshold of pressure detection is considered to be +/- 0.05 psig. A sample port on the bell jar assembly allows for gas sampling of the cavity space. Constant ambient test temperature has been achieved by using modified low cost temperature controlled insulated enclosures, readily available off the shelf.

Bell Jar Tech Specs:

Bell Jar Material:	304 Stainless Steel
Bell Jar Size (internal):	4.31 in. ID, 7.15 in. height
Data Logger:	Field Point Data Acquisition, Labview Software on laptop
Pressure Transducer:	PTX 6600, absolute pressure, 0.8% accuracy, detector threshold system +/- 0.5 psi
Thermocouple:	0.5°F accuracy
Temperature Enclosure:	+/- 1.0°F

II. TESTING AND PRODUCT CANS

Sufficient bell jar equipment is available at the Savannah River Site (SRS) to simultaneously test up to six product cans containing plutonium bearing material. Seventy eight cans have been tested to date.

Packaging configurations for plutonium bearing materials nominally consist of can/bag/can where the outer can is a crimp-sealed food can. Inner cans may be slip lid (tape sealed), screw top, or crimp-sealed cans. The bag (nominally polyethylene) is used for contamination control and may be heat sealed or tape sealed. In some cases filtered outer cans are used.

An implicit assumption in the bell jar test is the free flow of gas generated from its source through the inner can, bag, and outer can. Generated gas which can not leak out will pressurize internal containers. Pressurization of the outer can is readily detected using a lid deflection tool.

Cans are retrieved from storage and smeared for contamination prior to testing. All cans had less than 20 dpm/100 sq. cm alpha contamination when loaded into the bell jars. At the end of the test all cans were found to have less than 20 dpm/100 sq. cm alpha contamination and the bell jar inside surface was also found to have less than 20 dpm/100 sq. cm alpha contamination. There has been no evidence of loss of contamination control during the test.

The ambient test temperature is maintained by the constant temperature enclosure and heater controller. A temperature of 35^{o}C (95^{o}F) is typically used.

III. TEST RESULTS

Seventy eight cans containing plutonium oxide or plutonium contaminated enriched uranium oxide have been tested to date. Table 1 provides the length of the test, change in pressure, and lid deflection during the test.

Table 1a: Test Results

Item No.	Test Length (days)	Pressure Change (psig)	Lid Deflection (in.)
1	20	-0.10	N/A
2	20	0.18	N/A
3	20	0.00	N/A
4	15	0.04	N/A
5	15	-0.03	N/A
6	15	0.04	N/A
7	15	-0.04	N/A
8	15	-0.04	N/A
9	15	-0.01	-0.001
10	15	-0.02	0.001
11	15	0.00	0.006
12	15	0.05	0.016
13	15	0.01	0.006
14	18.5	-0.29	filter
15	18.5	0.12	0.032
16	26	-0.42	filter
17	26	-1.23	filter
18	12	-0.09	0.006
19	41	-1.91	-0.015
20	17	0.04	filter

The lid position was measured using a dial indicator micrometer, starting with item number 9, during loading and unloading of the bell jar. The change in position is computed as a deflection. A positive lid deflection indicates that the outer crimp sealed can pressurized (perhaps very slightly) during the test. Zero or nearly zero lid deflection coupled with a definitive change in pressure indicates a leaky outer can. Many of the cans tested have a filter embedded into the outer can lid (manufacture's lid). These cans are indicated with the word filter in the last column. The lid deflections recorded for items 15, 19, 21, and 22 are significant (greater than 10 mills) enough to indicate the outer crimp sealed can is at least partially sealed. For items 12 and 31 the significant lid deflections and the lack of pressurization indicates that the outer crimp sealed cans are well sealed.

Table 1b: Test Results

Item No.	Test Length (days)	Pressure Change (psig)	Lid Deflection (in.)
21	25	1.10	0.050
22	29	-1.67	-0.031
23	35	0.73	filter
24	17	0.07	0.005
25	17	0.08	0.006
26	9	0.01	-0.01
27	9	-0.15	filter
28	9	-0.05	filter
29	9	0.03	-0.003
30	9	-0.01	filter
31	8	0.01	0.012
32	8	-0.57	0.005
33	8	0.10	0.004
34	8	-0.22	filter
35	8	-0.08	filter
36	33	0.94	filter
37	7	-0.05	filter
38	7	-0.19	filter
39	7	-0.06	filter
40	7	-0.77	filter

Table 1c: Test Results

Item No.	Test Length (days)	Pressure Change (psig)	Lid Deflection (in.)
41	33	-0.52	filter
42	15	-0.04	-0.00
43	15	-0.01	0.000
44	15	-0.16	filter
45	15	0.00	0.003
46	14	0.89	0.000
47	14	0.14	filter
48	14	0.66	-0.009
49	14	0.11	-0.002
50	14	0.09	filter
51	14	0.06	filter
52	14	0.06	filter
53	14	-0.02	filter
54	14	-0.09	filter
55	8	-0.01	filter
56	8	-0.02	filter
57	8	0.13	filter
58	8	0.01	filter
59	8	0.00	filter
60	8	-0.01	filter

Table 1d: Test Results

Ite7m No.7	Test Length (days)	Pressure Change (psig)	Lid Deflection (in.)
61	7	0.28	filter
62	7	0.18	filter
63	7	-0.12	filter
64	7	-0.02	filter
65	7	0.01	filter
66	7	0.00	filter
67	11	-0.04	filter
68	11	0.08	filter
69	11	-0.10	filter
70	11	0.10	filter
71	11	-0.11	filter
72	11	0.04	filter
73	12	-0.62	filter
74	12	-0.13	filter
75	12	-0.07	filter
76	12	-0.11	filter
77	12	-0.35	filter
78	12	-0.17	filter

Items which have a pressure change greater than 0.01 psi per day (either positive or negative) are identified in Tables 2 and 3.

Table 2: Description of Ten Items Which Resulted In Significant Pressurization

Item No.	Description
21	Scrap unirradiated uranium/plutonium fuel, passivated plutonium-iron and uranium alloy, original can, 244 grams of plutonium
23	Same as number 21 except material re-packaged due to pressurizing inner can (bulging), 183 grams of plutonium
33	Enriched uranium oxide/carbide fuel, re-packaged 3/02, source is Westinghouse
36	Re-packaged item number 21
46	Sweepings standards comprised of a mixture of plutonium tetra-fluoride, plutonium oxide, and magnesium oxide. 376 grams of plutonium.
47	Same as number 46, 424 grams of plutonium.
48	Same as number 46, 108 grams of plutonium.
57	Plutonium oxide mixed with magnesium oxide, source is Rocky Flats, 32 grams of plutonium
61	Plutonium oxide mixed with magnesium oxide, source is Rocky Flats, 21 grams of plutonium
62	Plutonium oxide mixed with magnesium oxide, source is Rocky Flats, 24 grams of plutonium

Table 3: Description of Seventeen Items Which Resulted In Significant Depressurization

Item No.	Description
14	Plutonium and depleted uranium oxide, re-packaged 1/02, original crimp sealed can was inward deflecting
16	Same as item number 33.
17	Same as item number 16
19	SRS plutonium oxide, packaged 2/02, 700 grams of plutonium
22	SRS plutonium oxide, packaged 6/02, 669 grams of plutonium
27	Plutonium, enriched uranium and zirconium oxide fuel, source is Nuclear Material and Equipment Co. (NUMEC)
32	Same as number 16 except depleted uranium
34	Plutonium oxide with slightly enriched uranium oxide, source is Batelle
38	Plutonium and uranium oxide fuel, plutonium is made from oxalate precipitation and uranium is oxide is physically mixed, source is GE Vallecitos
40	Same as number 38
41	Unirradiated plutonium and uranium oxide fuel, 244 grams of plutonium, re-packaged twice due to signs of outer can pressurizing (bulging)
44	Same as number 38
63	Metal turnings, 867 grams of plutonium
73	Plutonium and enriched uranium oxide, high amounts of molybdenum, source is Argonne, 245 grams of plutonium
74	Plutonium and enriched uranium oxide, high amounts of carbon, source is Argonne, 134 grams of plutonium
77	Plutonium and enriched uranium oxide, high amounts of carbon, source is Argonne, 88 grams of plutonium
78	Plutonium and enriched uranium oxide, high amounts of carbon, source is Argonne, 148 grams of plutonium

The items identified with significant pressurization tend to have high levels of certain impurities such as magnesium oxide. Items 21 and 23 are known to have ammonium chloride which is very hygroscopic. Pressurization of number 33 is surprising since similar material resulted in depressurization (item 16). However, item 33 was tested for only 7 days and a longer test may indicate minimal pressurization (less than 0.01 psig per day). The source of pressurization for all items in Table 2 is most likely hydrogen. Hydrogen is generated by radiolysis of adsorbed moisture. The ability to predict those cans which lead to pressurization is complex, although the data presented suggests that magnesium oxide is very likely a contributor to pressurization.

The items identified with significant depressurization are both older and newly created materials. Depressurization is associated with plutonium oxides created using oxalate precipitation and high fired processes used to create fuels. The presence of uranium does not prohibit depressurization. The source of depressurization is very likely oxygen consumption. The exact mechanism is less clear although chemical reaction with the sub stoichiometric plutonium oxide has been theorized [1]. The test results presented in no way confirm or reject this theory.

IV. CONCLUSIONS

Bell jar pressurization test results for seventy eight items are presented. The items tested represent a very broad selection of plutonium and uranium bearing materials, most of which is scrap from suspended or deactivated processes. The bell jar testing is being used at SRS to verify that excessive pressurization inside a shipping container will not occur during transportation. In general, most plutonium materials consume gas and therefore cause a depressurization when sealed inside a leak tight vessel. Depressurization is observed for both newly created and older materials. The presence of magnesium oxide appears to be linked with pressurization, although the magnesium oxide is not a necessary impurity for pressurization. Lastly, the vast majority of crimp sealed food pack cans leak sufficiently to allow for pressurization or depressurization to occur inside a shipping container containment vessel.

V. ACKNOWLEDGMENTS

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VI. REFERENCES

1. DOE STANDARD, Stabilization, Packaging, and Storage of Plutonium Bearing Materials, DOE-STD-3013-2000.