THE EFFECT OF VIBRATION ON ALPHA RADIOLYSIS OF TRANSURANIC (TRU) WASTE

A. Zerwekh*, J. Warren**, and S. Kosiewicz
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

ABSTRACT

This paper reports on previously unpublished scoping work related to the potential for vibration to redistribute radionuclides on transuranic (TRU) waste. If this were to happen, the amount of gases generated, including hydrogen, could be increased above the undisturbed levels. This could be an important consideration for transport of TRU wastes either at DOE sites or from them to a future repository, e.g., the Waste Isolation Pilot Plant (WIPP). These preliminary data on drums of real waste seem to suggest that radionuclide redistribution does not occur. However, improvements in the experimental methodology are suggested to enhance safety of future experiments on real wastes as well as to provide more rigorous data.

BACKGROUND INFORMATION

Alpha particles interact with hydrogenous materials to generate gases. This process has been studied for a considerable period of time at the Los Alamos National Laboratory (LANL) as well as at other DOE sites. At LANL, we have previously reported studies on the effects of alpha radiolysis of transuranic (TRU) radionuclides on different waste materials in both laboratory-scale vessels and actual drums of TRU waste. Extensive laboratory studies were reported by Zerwekh (1) and Kosiewicz (2), while drum scale work on real TRU wastes was reported by Zerwekh and Warren (3) and Warren and Zerwekh (4).

Decreases in the rates of gas production have been documented as the total radiation doses deposited in the waste matrices increased. This is attributed to matrix depletion within the limited range of alpha particles emitted from the radionuclide particles. Figure 1 shows deterioration of cellulosic matrices resulting from various levels of radionuclide contamination. Some of the degraded materials appear to be charred.

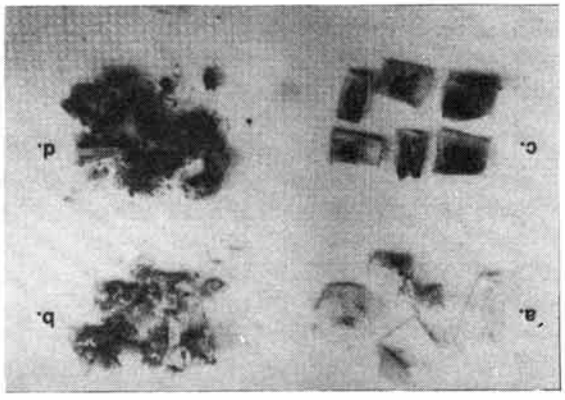

Fig. 1. Radiolytically degraded cellulosics. (a) Paper after 24 months exposure at a dose rate of 4×10^4 nCi/g. (b) Paper after 19 months exposure at a dose rate of 400×10^4 nCi/g. (c) cotton cloth after 24 months exposure at a dose rate of 40×10^4 nCi/g. (d) Cotton cloth after 18 months exposure at a dose rate of 3200×10^4 nCi/g.

A measure of the radiolytically generated gases is the G(gas) value. The definition of G(gas) is the number of total gas molecules evolved for each 100 eV of energy deposited in a waste matrix. For alpha radiolysis caused by radionuclides embedded in particles, we recommend that G(gas) should not be considered to be an intrinsic property of the material studied because it is also a function of the intimacy of contact between the waste matrix and the TRU contaminant. During the course of the studies, G(gas) is not significantly effected by changes in the activity levels of the radioisotopes because they have relatively long half lives compared to the durations of the studies.

For cellulosics, Kosiewicz (2) showed that the G (gas) value decreased asymptotically from an initial value of about 1.9 gas molecules per 100 eV deposited in the matrix to a value of about 0.5 when these particular experiments were terminated (Fig. 2).

EXPERIMENTAL PROCEDURE

For the currently reported work, we retrieved six drums of plutonium-238 contaminated waste from storage to study during 1984 and 1985. The drums consisted of either mixed cellulosic wastes or mixed combustibles as indicated in Table 1. They ranged in age from four to ten years old. Four of these drums were relatively undisturbed, while two of them were tumbled in a drum tumbler for four hours. We propose that end-over-end tumbling is a more vigorous agitation than would be anticipated for normal handling or transportation. Evolved gases were analyzed by mass spectrometer for hydrogen and other constituents.

The wastes were contained in 30-gallon drums which were stored in concrete casks buried retrievably. None of the drums had filters in them or were vented, other than that which might occur through the lid gasket. Upon retrieval, the 30-gallon drums were penetrated and gas samples were taken immediately. A second gas sample was taken just prior to perforating the lid of the 30-gallon drum and placing it into 55-gallon overpack drums. From this point on, the gas samples were taken from the headspaces of the 55-gallon drums. $G(H_2)$ values were estimated from these latter analytical data.

* Retired LANL consultant.
** Now with NFT, Inc., Lakewood, CO.

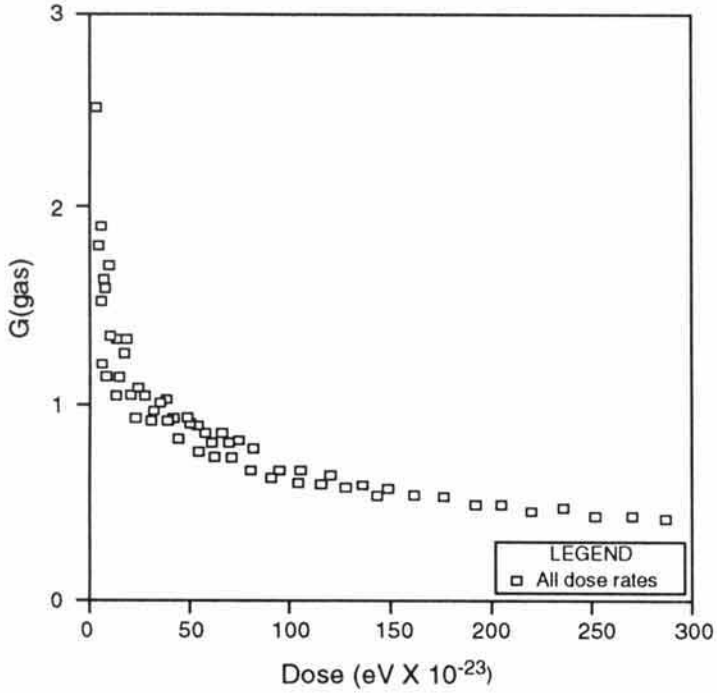

Fig. 2. Decrease in G(gas) with increasing dose. For cellulosics.

RESULTS

The $G(H_2)$ values of the first set of drums were estimated to be 0.023, 0.007, 0.002, and 0.007 (Table II). This compares to those of 0.017 and 0.139 measured for the tumbled set of drums. All of these $G(H_2)$ values measured on real TRU wastes corroborate the matrix depletion observed for the laboratory-scale experiments. While the last number (0.139) is the largest one currently measured, it is still smaller than $G(H_2)$ values of 0.2 - 0.4 (Table III) that were measured on newly generated drums of somewhat similar waste as reported by Warren and Zerwekh (4). In addition, there was a high level of hydrogen measured in this drum prior to its tumbling. Its initial hydrogen concentration upon retrieval was 4.3 mol%. When the study was terminated five months later, the hydrogen had increased to 11.9 mol%, while the oxygen concentration was 13.0 mol%. This is well into the region of hydrogen deflagration and possibly detonation. None of the other drums studied showed hydrogen concentrations that exceeded the lower flammability limit.

It should be noted that the newly generated TRU wastes were not exactly the same type of wastes as those used for the tumbling study. Different materials have been shown (5) to have corrected G(gas) values ranging from 0.04 to possibly as high as 6.3. Consequently, one might expect somewhat different levels and types of gas to be produced from different waste

TABLE II
$G(H_2)$ Values

Drum	Tumbled	G(H2) Values
BFB-7	no	0.023
BFB-98	no	0.007
BFB-97	no	0.002
BFB-99	no	0.007
BFB-21	yes	0.017
BFB-24	yes	0.139

TABLE I
Waste Drums

Drum ID	Age (years)	^{238}Pu Loading (gm)	^{238}Pu Loading (alpha Ci)	Waste Type LANL code	Waste Type Matrix
BFB-7	4	5.6	78	A-60	other combustibles
BFB-98	10	26.1	365	A-15	mixed cellulosics
BFB-97	10	11.8	165	A-15	mixed cellulosics
BFB-99	10	36.6	512	A-15	mixed cellulosics
BFB-21	4	4.8	67	A-60	other combustibles
BFB-24	4	7.1	99	A-60	other combustibles

TABLE III
Comparison Drum G Values*

Drum ID	^{238}PuLoading (gm)	^{238}PuLoading (alpha Ci)	Waste Matrix	G(gas)	$G(H_2)$
BFB-112	1.2	16.8	plastic, lead gloves	0.49	0.20
BRB-114	15.6	218	rags & plastics	0.86	0.40
BFB-118	4.9	69	rags, plastics, metals	0.54	0.27

*Drums of newly generated waste

matrices. For example, plastics produce more hydrogen than cellulosics for a given level of radionuclide contamination.

In the laboratory-scale experiments (1,2), test vessels were vigorously shaken manually, and no increases in the gas evolution rates were observed. However, there is a minor possibility that the test containers for these tests did not have sufficient space to allow the radionuclides to redistribute. Taken in concert, the data from the drum and laboratory work appear to suggest that the radionuclide contaminant is not significantly redistributed upon vigorous tumbling. It is speculated that redistribution does not occur because charred waste matrix may partially encapsulate the radionuclide particulates. Consequently, fresh matrix is not brought within the limited range of alpha particles emitted from the radionuclide particulates.

SUGGESTED IMPROVEMENTS FOR FUTURE WORK

There are improvements that can be made to these initial experiments to enhance safety as well as data interpretations. First, some of the drums were initially found to have potentially flammable mixtures of hydrogen and oxygen. We recommend replacing the ambient gases in the drums with an inert gas to prevent either a hydrogen deflagration or detonation. We have previously shown (2) that this does not effect the G values determined. Second, if, during the course of the study, the hydrogen concentration exceeds the lower flammability limit, the experiment should be terminated and the gases in the headspaces of the drums replaced with an inert gas. Third, initial penetrations of the retrieved drums should be done by a device contained in an explosion containment vessel. Such a drum venting system is in the initial design stages at LANL (6).

To enhance data interpretations, *first*, we recommend that the G values be determined on all experimental drums. Then, the drums should be tumbled, and the G values redetermined for comparison to the initial numbers on the same drums. Second, all comparison data should be done on drums of the same waste matrix type. Finally, surface examination of the depleted wastes by Auger spectroscopy, ESCA, or possibly electron microscopy may provide direct evidence that the radionuclide particles have been partially encapsulated in charred waste matrix.

REFERENCES

1. A. ZERWEKH, "Gas Generation from Radiolytic Attack of TRU-Contaminated Hydrogenous Waste," Los Alamos National Laboratory report LA-7674-MS (June 1979).
2. S.T. KOSIEWICZ, "Gas Generation from Organic Transuranic Wastes. I. Alpha Radiolysis at Atmospheric Pressure," Nucl. Techn. V54, pp. 92-99 (July 1981).
3. A. ZERWEKH and J.L. WARREN, "Gas Generation and Migration Studies Involving Recently Generated 238Pu-Contaminated Waste for the TRU Waste Sampling Program," Los Alamos National Laboratory report LA-10732-MS (July 1986).
4. J.L. WARREN and A. ZERWEKH, "TRU Waste Sampling Program," Los Alamos National Laboratory report LA-10479-MS (July 1985).
5. S.T. KOSIEWICZ, comments on "Gas Generation from Organic Transuranic Wastes. I. Alpha Radiolysis at Atmospheric Pressure," Nucl. Techn. V.92, p. 428 (Dec. 1990).
6. S.T. KOSIEWICZ, Los Alamos National Laboratory, personal communication (November 1992).