HYDROGEN EXPLOSION TESTING WITH A SIMULATED TRANSURANIC DRUM (U)*

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ABSTRACT

Transuranic (TRU) waste generated at the Savannah River Site (SRS) is currently stored onsite for future retrieval and permanent disposal at the Waste Isolation Pilot Plant (WIPP). Some of the TRU waste is stored in vented 210-liter (55-gallon) drums and consists of gloves, wipes, plastic valves, tools, etc. Gas generation caused by radiolysis and biodegradation of these organic waste materials may produce a flammable hydrogen-air mixture (>4% v/v) in the multi-layer plastic waste bags. Using a worst case scenario, a drum explosion test program was carried out to determine the hydrogen concentration necessary to cause removal of the drum lid. Test results indicate an explosive mixture up to 15% v/v of hydrogen can be contained in an SRS TRU drum without total integrity failure via lid removal.

INTRODUCTION

The Savannah River Site (SRS), which is operated for the U.S. Department of Energy by Westinghouse, produces nuclear materials for national defense, scientific research, and medical applications. TRU waste generated during routine operation is disposed of in polyvinylchloride (pvc) and polyethylene bags and sealed using a "horsetail" technique. Waste cuts are contained in multiple bag layers for contamination control purposes and placed inside a high-density polyethylene drum liner. The vented liners are loaded into vented 55-gallon (210 liter) galvanized steel drums and placed on concrete pads for interim storage and future retrieval.

Radiolysis causes bag degradation, which may produce enough hydrogen to exceed the lower flammability limit of a gas-air mixture in the bags or liner. To prevent hydrogen buildup in the drum and liner, both of the lids have carbon composite filter vents that allow hydrogen to escape by natural diffusion. This filter vent, however, does not prevent the possibility of a flammable hydrogen-air mixture forming in the individual waste cut bags.

Tests to determine the hydrogen-air concentration required to cause a waste drum integrity failure (drum lid removal) were conducted at the Du Pont Explosion Hazards Laboratory, Remote Test Site, Chambers Works, NJ.

TEST PLAN

Testing took place in three segments

- 1.7-liter pressure vessel tests
- drum hydrogen-air mixing tests
- drum explosion tests

The primary test objective was to determine the hydrogen concentration that was capable of causing a drum integrity failure during an explosion. A failure was defined as lid removal. An additional test objective was to determine the maximum pressure and the rate of pressure rise versus hydrogen concentration for a 1.7-liter and a 55-gallon vessel.

Pressure Vessel Test

Hydrogen was added to a 1.7-liter vessel, a Nixon reactor, using partial pressure addition. According to Dalton's law of partial pressure,

$$P_1/P_t = n_1/n_t = \text{mole fraction} = y_1, \qquad \text{(Eq.1)}$$

where

p_t = total pressure,

p_1 = partial pressure of component 1,

n_1 = moles of component 1,

n_t = total moles.

Therefore, a vessel at barometric pressure can be filled to a specific gas concentration by measuring the total gas pressure. Use of this relationship assumes hydrogen behaves as an ideal gas. Once the Nixon reactor equilibrated to barometric pressure, hydrogen was added through a valved inlet to the selected concentration using this procedure. A homogeneous hydrogen-air mixture was attained by agitation for ten minutes. Data acquisition devices recorded the maximum pressure and pressure rise during the gas mixture ignition.

The Nixon reactor (Fig. 1) was equipped with a thermocouple, a pressure transducer, an ignitor, an agitator, and a valved gas inlet. A transducer and a thermocouple relayed temperature and pressure information to a data acquisition system.

For this test, the Nixon reactor was filled to slightly above ambient pressure with known hydrogen concentra-

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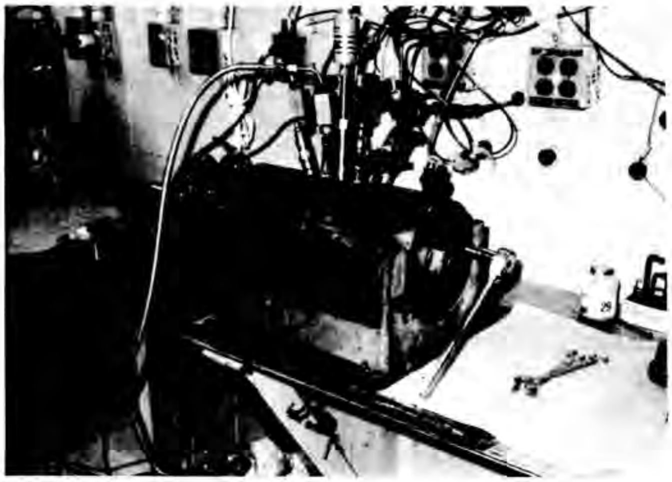

Fig. 1. Nixon Reactor.

tions (5-50% v/v) and ignited to determine the maximum pressure and rate of pressure rise. Using this data as a baseline measurement for the drum explosion tests, the concentration range of interest was selected by noting any steep increase in the maximum pressure of the vessel and the rate of pressure rise. These baseline measurements could also be used to make a comparison between the Nixon reactor and the drum explosion data.

Drum Mixing Tests

A mixing test was conducted using two different hydrogen-air mixtures (5% and 25% v/v of hydrogen) to determine the natural diffusion time necessary for obtaining a homogeneous gas mixture in the drum. The mixing drum was a standard SRS-galvanized TRU drum containing a polyethylene liner with carbon-media filter vents in the drum and the liner lids. A modified filter element allowed the drum vent to be plugged during testing and three ports along the drum side were used for gas sampling (Fig. 2).

Hydrogen concentrations for all drum tests were obtained by evacuating the drum prior to adding hydrogen. This procedure was necessary to ensure that there would be no hydrogen leakage from the drum during equilibration and to simulate the TRU pad drum conditions. Gas samples were drawn every 20-30 minutes from three different sampling ports and analyzed using gas chromatography. The sampling ports were 1/8-inch steel pipes that extended 51 mm (2 inches) from the drum side. They were welded to the drum and sealed to the liner. Septa were attached to the drum ports and gas samples (10 cc) were drawn from the drum using plastic syringes. To ensure that there was no measurable gas diffusion when using the syringes, each one was subjected to a qualification test. The test consisted of filling each syringe with a 10% v/v hydrogen standard and allowing it to sit for 30 minutes before injection into the gas

Fig. 2. Sampling ports for the drum mixing tests.

chromatograph. No measurable hydrogen loss out of the syringes was detected.

Drum Explosion Tests

The final testing segment was to determine the concentration necessary to cause a TRU drum lid removal during a hydrogen explosion. To perform drum testing, a 1/2-inch

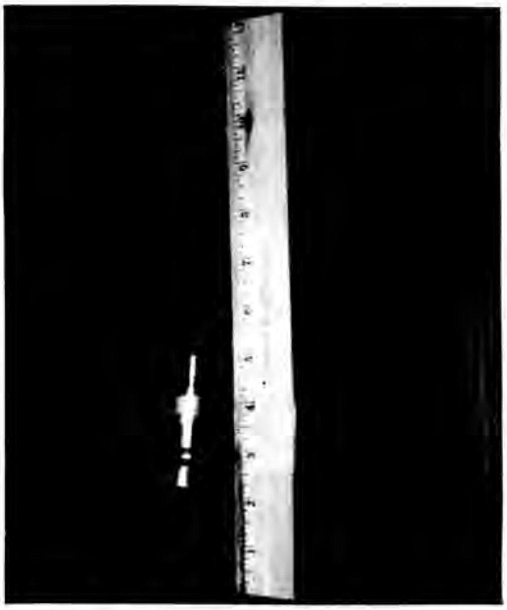

Fig. 3. Modified filter vent.

Fig. 4. Explosion test drum equipment setup.

steel pipe that extended 51 mm (2 inches) was added to the standard filter vent (Fig. 3).

The pipe extension allowed the vent to be opened or closed during the drum evacuation and filling processes. The explosion test drums were fitted with a 2-inch diameter steel pipe that was located midway along the drum height. This pipe was used for the insertion of ignition and data acquisition devices (Fig. 4).

This pipe was sealed to the galvanized drum and the polyethylene liner by welding it to the drum and using a lock nut and sealant at the pipe-liner interface. Ignition and data acquisition devices were sealed to the drum using a modified flange. The flange had access ports for a pressure transducer, a thermocouple, a Nichrome™ ignitor, and gas inlets.

During explosion testing, the drum was evacuated via a vacuum line at the flange-drum interface and filled with a hydrogen concentration that ranged between 12 and 36% v/v. The filter vent was plugged and the gas mixture was allowed to equilibrate by natural diffusion. Prior to igniting the gas mixture with a hot-wire device, a gas sample was drawn into a 1-liter Hoke to verify the hydrogen concentration using gas chromatography. After ignition, the pressure, the rate of pressure rise, and any additional observations were recorded.

TEST RESULTS

Pressure Vessel Testing

Maximum pressure and pressure rise per unit time were highly dependent on hydrogen concentration. Fig. 5 is a graphical representation of the empirical relationship established from the pressure vessel data. A third-order polynomial gives a good fit for the figure with a correlation

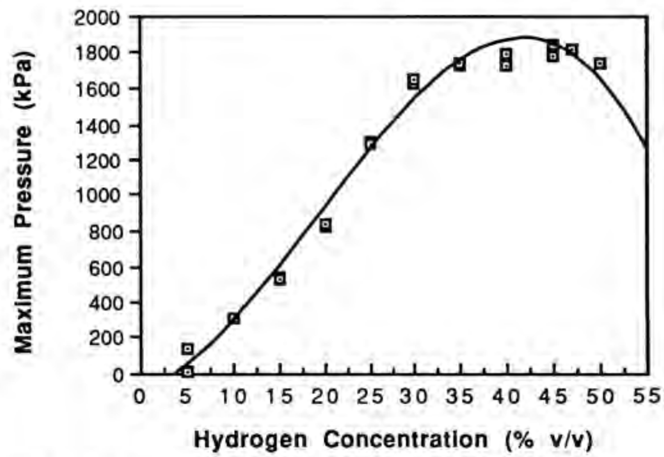

Fig. 5. Nixon reactor maximum pressure vs. hydrogen concentration.

coefficient of 0.99. A similar relationship was also obtained for the rate of pressure rise as a function of time.

Drum Mixing Tests

Initially, partial pressure addition was attempted in order to obtain various hydrogen concentrations, but severe leaking at the drum-lid interface and excessive deformation of the drum lid prevented this approach. To prevent any future lid deformation and to eliminate leaks at the drum-lid interface, drums were evacuated prior to hydrogen addition to achieve concentrations of 5% and 25% v/v. Testing indicated that a 25% v/v hydrogen-air mixture produced a significant gas layering effect for the first 10-30 minutes (Fig. 6). Based on these results, 50 minutes was chosen as an adequate equilibration time for the drum explosion tests.

Simulated TRU Drum Explosion Tests

Hydrogen concentrations in the explosion tests were obtained by applying the same evacuation method as used in the drum mixing tests. The test data are shown in Table I. During the first eight tests, a successful reaction was only achieved twice. Although 64 mm (2.5 inches) of 20-gauge Nichrome ™ wire had been used in the pressure vessel tests, 114 mm (4.5 inches) of wire was substituted for the drum explosion tests. This additional wire length effectively increased the resistance and reduced the wire heat load, which resulted in a cooler resistor and prevented the gas mixture from reaching its auto-ignition temperature. Reduction of the wire length to 76 mm (3 inches) resulted in successful reactions for all tests except No.15. Equipment inspection showed a high probability that the ignition source was shorted out by a thermocouple that was resting against the area where power was supplied to the drum. Repositioning and insulation of the thermocouple prevented any reoccurrence of this problem.

Supplying hydrogen to the drums using partial pressure addition frequently proved to be within approximately 2% of the desired concentration. The actual concentrations reported in Table I are the averages of the gas chromatography readings taken prior to ignition. Standard deviations have been reported whenever more than one sample could be drawn and successfully analyzed. Due to an integrator-area calculation error, the values presented for the hydrogen concentration and standard deviation in Test 16 were hand-calculated. This was accomplished by comparing the peak area mass of an unknown chromatograph concentration to that of a known gas standard chromatograph. Uncertainty analysis calculations account for the large variation in the standard deviation compared to the other test runs.

The maximum pressure as a function of hydrogen concentration for all tests is shown in Fig. 7. Figure 7 follows a well-defined pattern, except for the point at 14% hydrogen. Since the point cannot be dismissed as an outlier, additional testing is required to verify any functional relationship.

Observations made during each test showed that removal of the drum lid occurred at concentrations of 35%, 22%, 18%, and 17%. In the five successful tests at less than 17%, only bulging of the drum top and bottom occurred. Since a limited number of data points could be obtained, a statistical probability for drum failure as a function of hydrogen concentration could not be ascertained.

CONCLUSIONS

The data suggest that an explosive mixture up to 15% v/v of hydrogen can be contained in a 210-liter (55-gallon) TRU drum without total integrity failure via lid removal. An empirical relationship was defined for the pressure and pressure rise as a function of hydrogen concentration for an experimental pressure vessel. Similar empirical relationships could not be established for the drum explosion tests. This was due to the limited number of tests and drum closure variables. No direct correlation could be made between the explosion and pressure vessel test data.

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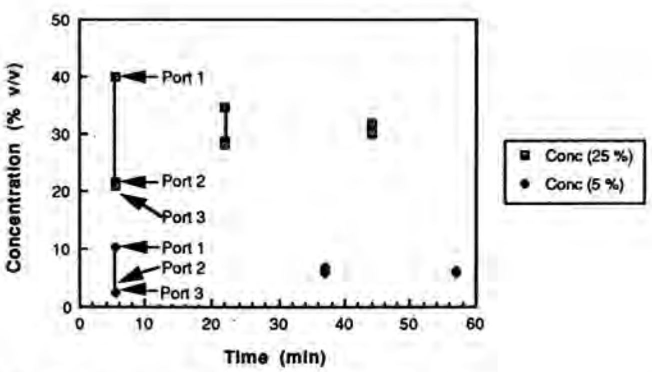

Fig. 6. Hydrogen concentration vs. time.

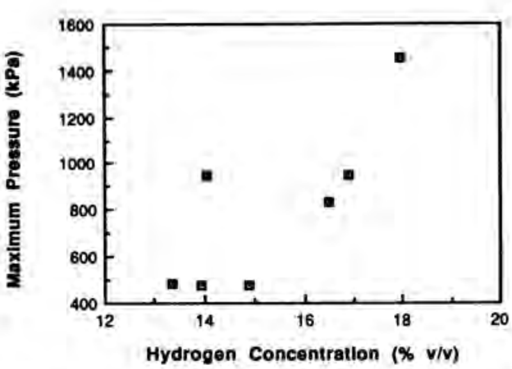

Fig. 7. Maximum pressure vs. hydrogen concentration.

TABLE I

Drum Explosion Data

Test No.	Filled H_2 Concentration (%)	Actual H_2 Concentration (%)	Standard Deviation (±)	Maximum Pressure (kPa)#	dP/dt Maximum (kPa/s)#	Observations
1†	15	N/A	N/A	N/A	N/A	none
2	15	14.61	N/A	N/A	N/A	none
3	25	N/A	N/A	855	68 300	lid blown
4	20	12.843	0.604	644	159	flame propagation
5	20	13.594	1.283	N/A	N/A	none
6	25	28.219	0.134	N/A	N/A	none
7	25	28.708	0.632	N/A	N/A	none
8	20	18.806	1.572	N/A	N/A	none
9††	20	35.329	0.458	725	106 000	lid blown
10	15	16.95	N/A	948	99 790	lid blown
11	15	14.893	0.098	476	3902	bulged
12	15	14.053	0.021	952	97 550	bulged
13	15	6.49	0.706	837	261 800	bulged
14	12	13.346	0.253	485	122 700	bulged
15†††	18	17.549	0.429	61	6.9	none
16*	18	22.72	3.04	2206	686 900	lid blown
17	18	17.966	1.094	1455	491 600	lid blown
18	12	13.945	0.277	476	87 480	bulged

gage pressure readings
† 114-mm Nichrome™ wire used for ignition (Tests 1-8)
†† 76-mm Nichrome™ wire used for ignition (Tests 9-18)
††† thermocouple shorted out power source
* calculated concentration based on chromatography peak mass
N/A not available