Preprint of a paper published in *Journal of Nuclear Materials Management*, XXXVIII(3):43-53, 2010. Also available as SRNL-STI-2010-00053

Pressure Integrity of 3013 Container under Postulated Accident Conditions

George B. Rawls, Jr., Savannah River National Laboratory; F. Coyne Prenger, Los Alamos National Laboratory; Joseph E. Shepherd, California Institute of Technology; Zhe Liang, Atomic Energy of Canada Limited – Chalk River Laboratories.

Abstract

A series of tests was carried out to determine the threshold for deflagration-to-detonation transition (DDT), structural loading, and structural response of the Department of Energy 3013 storage systems for the case of an accidental explosion of evolved gas within the storage containers. Three experimental fixtures were used to examine the various issues and three mixtures consisting of either stoichiometric hydrogen-oxygen, stoichiometric hydrogen-oxygen with added nitrogen, or stoichiometric hydrogen-oxygen with an added nitrogen- helium mixture were tested. Tests were carried out as a function of initial pressure from 1 to 3.5 bar and initial temperature from room temperature to 150 °C. The explosions were initiated with either a small spark or hot surface. Based on the results of these tests under the conditions investigated, it can be concluded that DDT of a stoichiometric hydrogen-oxygen mixture (and mixtures diluted with nitrogen and helium) within the 3013 containment system does not pose a threat to the structural integrity of the outer or ultimate containment.

Introduction

A system composed of triple-nested stainless steel 3013 storage canisters used to store plutonium-bearing powders was evaluated to determine the probability of plutonium bearing material release in the event of a hydrogen-oxygen explosion. Generation of hydrogen and oxygen within the storage containers raises the possibility of internal combustion in the highly unlikely event of an ignition source being present. The 3013 Containment System contains no identifiable ignition source. However, because hydrogen has a very low ignition energy, a concern was raised that friction-generating events caused by the interaction of the nested containers may have the potential to provide sufficient energy to ignite a combustible hydrogen-oxygen mixture.

The California Institute of Technology, Explosion Dynamics Laboratory (Liang and Shepherd 2007) was contracted to perform a series of tests designed to evaluate the potential for detonation and the resulting structural response of the 3013 container system. This containment system is used complex wide to package plutonium metal and oxides under the Department of Energy (DOE) 3013 Packaging Standard. An illustration of the 3013 containers is shown in Figure 1. The convenience and inner containers used at each DOE facility differ but the 3013 outer containers are the same for all configurations throughout the DOE complex.



Figure 1 Nested 3013 containers. The outer container is on the left, the inner container in the middle, and the convenience container on the right.

The test program used deliberate ignition of explosive mixtures to determine the type of explosion, (i.e. deflagration having a subsonic burn front or a detonation having a supersonic burn front), structural loading (pressure history), and structural response (strain history) in both simulated test fixtures and actual 3013 outer containers.

Testing Methodology

The initial gas pressures, temperatures and gas compositions in the storage containers were based on the surveillance data for the storage material containers of interest. The test fixtures and explosive mixtures were designed to address all the identifiable modes of explosions possible in the 3013 storage system. The 3013 outer container was treated as the final containment barrier and any structural benefit derived from the inner containers was neglected, for added conservatism, in the final evaluation.

Tests were carried out in three parts to determine the threshold for DDT, structural loading, and structural response of the nested storage canisters. Three experimental fixtures were used to examine the various issues.

The first series of tests was performed to understand the influence of the small gaps between the inner and outer containers on DDT. Because the ratio of gap thickness between the outer and inner containers to container diameter was small, a planar fixture was used to simulate the combustion behavior. Because of the planar configuration, only the pressure time history was measured in these tests. A second series of tests was performed in a thick-wall cylindrical container fitted with a cylindrical insert to simulate the outer container-inner configuration. In this cylindrical geometry, except pressure measurement, strain gages were used effectively to measure the structural response of the thick-walled outer container. In addition, the eccentricity in the annular gap between outer and inner cylinders was also investigated. The final test fixture was an actual 3013 outer container modified with penetrations for pressure transducers, gas handling, and ignition sources.

Three gas mixtures, chosen to bound the anticipated container gas compositions, consisting of a stoichiometric hydrogen-oxygen mixture (Mixture A), a stoichiometric hydrogen-oxygen mixture with added nitrogen (Mixture B) or a stoichiometric hydrogen-oxygen mixture with an added nitrogen- helium mixture, (Mixture C) were tested. Tests were carried out as a function of initial pressure ranging from 1 to 3.5 bar and initial temperature from room temperature to 150°C. The explosions were initiated with either a small spark or a glow plug (hot surface).

Planar Gap Tests

The planar gap test fixture was designed to determine the threshold for DDT in the storage system annular gaps. When the containers are nested, annular spaces are created between the various container walls and gaps are also formed between the container lids, as shown in Figure 2. The tests were carried out in a planar geometry simulating the annular gaps between the outer and inner containers of the 3013 storage system. The test fixture consisted of a pair of rigid flat plates with an adjustable gap between them representing the annular space between the nested storage system containers. Figure 3 shows a drawing of the planar test fixture showing the location of the pressure transducers. The gap was filled with a representative explosive gas mixture, ignited, and the subsequent explosion development was monitored using pressure transducers. For each mixture composition, the threshold for DDT was determined by varying initial pressures. Because the inner and outer containers could be eccentric, gap size was treated as a parameter, and values of 0.01", 0.02", 0.05", 0.1", 0.44" (0.254, 0.508, 1.27, 2.54, 11.18 mm) were investigated. The annular gap between the containers comprising the storage system could vary from 0 to 0.185" (0-4.7 mm) depending on the eccentricity of the containers. The largest gap represented the headspace gap of approximately 0.5" (12.7 mm).



Figure 2 A close-up drawing of an inner container nested within an outer container showing the headspace gap.



Figure 3 Planar fixture assembly 1-bottom plate, 2-top plate, 3-pressure transducer holes, 4-spacer SP-spark plug, GP-glow plug

The planar tests showed that all three mixtures would undergo DDT with threshold initial pressures between 1 and 3 bar. Typical test results representing gap sizes between

0.1" and 0.44" and the three mixtures tested at room temperature are shown in Figure 4. The nomenclature used in Figure 4 for the pressure traces is as follows: the data points are the maximum measured pressures at P1, P2, P3 and P4 respectively, P_{CV} is the constant volume explosion pressure, P_{CJ} is the Chapman-Jouguet (CJ) pressure and P_{CJref} is the calculated reflected CJ pressure. The values of P_{CV} , P_{CJ} and P_{CJref} were calculated using a chemical equilibrium program of Reynolds (1986) with realistic thermochemical properties.

The results illustrated in Figure 4 show that mixture A is the most sensitive to initial pressure and gap width, providing the lowest DDT threshold pressure, mixture B is intermediate, and mixture C is the least sensitive, providing the highest DDT threshold pressure. The smaller the gap size, the lower the threshold pressure for DDT. Again, as seen in Figure 4 for mixture A, DDT was observed at an initial pressure of 1.25 bar for a gap of 0.44" (11.18 mm) and 0.9-1.0 bar initial pressure for a gap of 0.1" (2.54 mm).

The effect of the initial temperature was also examined with the planner fixture. It was found that the DDT transition occurred at slightly higher initial pressure for all three mixtures when the initial temperature was increased to 150°C, and the peak pressure was lower. The tests with the other configurations were only performed at room temperatures (more conservative).

Thick-Walled Cylinder Tests

The thick walled tube was fitted with a solid cylindrical insert to simulate the annular gap between the outer and inner containers. Figure 5 provides a drawing the thick-walled test fixture showing the location of the pressure transducers and strain gauges. The positions of the strain gauges do not correspond to the positions of the pressure transducers. Three types of tube configurations were used: (1) empty tube (no insert), (2) tube with a concentrically located cylindrical insert, and (3) tube with an eccentrically located cylindrical insert. The gap configurations (2) and (3), together with the empty tube configuration (1), were chosen to cover the entire range of anticipated configurations in the 3013 storage system geometries. The test fixture was filled with one of the three representative explosive gas mixtures (A, B or C), ignited with a lowenergy spark, and the subsequent explosion development monitored with pressure transducers and strain gages. For each mixture composition and tube configuration, the threshold for DDT and corresponding structural response was determined for various initial pressures. Use of the thick-walled test fixture allowed all tests to be conducted with a single, fully instrumented test fixture because the deformations in each test remained in the elastic range.



Figure 4 Comparison of peak pressures for gap size 0.44-in and 0.10-in. The shaded region is the estimated threshold for the onset of DDT.





Thick-Walled Cylinder Tests; Configuration 1 (Empty Tube)

As demonstrated in the planar tests, the DDT threshold shifted to higher initial pressures for larger gap sizes. Tests with an open cylinder (Configuration 1) had no gap present and the highest DDT threshold pressures for all the gas mixtures were observed. Figure 6 shows the peak pressures and strains for mixture A. The DDT transition threshold was observed at an initial pressure of 2.5-2.6 bar for mixture A, and is twice as large as the DDT threshold, initial pressure of 1.2-1.25 bar for the largest gap size of 0.44" in the planar fixture (Figure 4a). All the transitions occurred near the tube end. The maximum strain was 170 microstrain at an initial pressure of 3.5 bar. For mixtures B and C, no DDT transition was observed in the empty tube for initial pressures up to 3.5 bar. In the 0.44" planar fixture, DDT was observed at an initial pressure of 2.1 bar for mixture B and 2.75 bar for mixture C (Figures 4c and e).



Figure 6 Peak pressures and strains for mixture A of thick-walled cylinder tests, configuration 1.

Thick-Walled Cylinder Tests; Configuration 2 (Concentric Insert)

The annular gap between inner and outer containers of the 3013 storage system varies between 0" and 0.16" (0–4.06 mm) depending on the eccentricity of the containers. The gap between the lids of the containers varies from 0.375" to 0.6" (9.5–15 mm) depending on the inner container cut-off length. For the tests an average annular gap of 0.08" with an average end gap of 0.5" was used in the thick-walled cylinder with concentric insert tests. A solid circular bar was inserted concentrically into the outer tube to create this geometry.

As shown in Figure 7, the DDT transition occurred at an initial pressure of 1 bar for mixture A. The DDT threshold was close to 1.5 bar for mixture B and 2.0 for mixture C.. The peak strains were always observed on the strain gauge close to the tube end , and the maximum value was 100 microstrain at an initial pressure of 3.5 bar.



Figure 7 Peak pressures and strains for mixture A of thick-walled cylinder tests, configuration 2.

Thick-Walled Cylinder Tests; Configuration 3 (Eccentric Insert)

In configuration 3, the more realistic case of inner container eccentricity was examined. In this test series, the solid cylinder inside the test cylinder was mounted eccentrically. The nominal minimum gap was 0.01" and maximum gap was 0.15". A diagram of the two configurations is shown in Figure 8.



Figure 8 Diagram of the two eccentric configuration thick-walled fixture assemblies.

In contrast to configuration 2, the annular gap size for configuration 3a (Figure 8a) was reduced on the pressure transducer side; therefore, one would expect faster DDT transition on this side. As shown in Figure 9, DDT indeed occurred right away at an initial pressure of 1 bar for mixture A, but the maximum strain was on the same order as the values recorded in configuration 2.

In configuration 3b (Figure 8b), the solid bar was rotated 180 deg, therefore, the largest gap, 0.15 in, appeared on the pressure transducer side, and the smallest gap was on the strain gauge side. For mixture A with an initial pressure of 1 bar, DDT appeared near the last transducer, P4 with configuration 3b but it was near the first transducer P1 with configuration 3a. This means that DDT occurred earlier on the smaller gap side and later on the larger gap side. This is consistent with the previous findings about the effect of the gap size on DDT thresholds in the planar fixture. As shown in Figure 10, there are no significant differences in the peak pressures and strains for the two configurations.



Figure 9 Peak pressures and strains for mixture A of thick-walled cylinder tests, configuration 3a.



Figure 10 Peak pressures and strains for mixture A of thick-walled cylinder tests, configuration 3b.

Calculated Pressures and Strain

The values for CJ pressure (P_{CJ}), reflected CJ pressure (P_{CJref}) and constant volume explosion pressure (P_{CV}) for each test were calculated using a chemical equilibrium program of Reynolds (1986) with realistic thermochemical properties.

The static strains, ε_{CJ} , $\varepsilon_{CJ ref}$, ε_{CV} , corresponding to the CJ, reflected CJ and constant volume explosion pressures, were inferred from the approximate stress-strain relation for a uniformly, statically loaded tube

$$\varepsilon = \frac{(P - P_a)R}{Eh} \tag{1}$$

where ε , E, R, h and P_a are strain, Young's modulus, average radius (R=(ID+h)/2), thickness of the tube, and atmospheric pressure, respectively.

Dynamic Load Factor

One of the most frequently used methods (Biggs, 1964, Paz and Leigh, 2004) to evaluate structural response to transient loads is the use of a Dynamic Load Factor (DLF). This method uses the measured or calculated peak pressure of the transient load corrected by the DLF to compute a static response, which has an equivalent deflection to the peak transient response. This method is useful if the dynamic load factor and peak pressure can be readily computed for the cases of interest.

The peak value of the strain signals can be analyzed by finding the DLF Φ , which is defined as the ratio of the measured peak strain to the peak strain expected in the case of quasi-static loading

$$\Phi = \frac{\varepsilon_{\max}}{\frac{\Delta PR}{Eh}}$$
(2)

The pressure term (ΔP) in Equation 2 can be based on either the measured peak value from the experimental measurement or one of the computed pressure values. Using the experimental pressure allows an evaluation of what type of loading (impulsive, sudden or mixed) is taking place. For an ideal single-degree of freedom structure and a simple pressure-time history with a single step function followed by a monotonic decay (Paz and Leigh, 2004, Biggs, 1964), values of DLF close to two are associated with the limit of "sudden loading" in which the pressure jumps to a high value and does not significantly decay on the time scale of the tube radial oscillation (breathing) period. In this regime, the peak elastic deformation is proportional to the peak pressure. As the decay time of the pressure after the step change becomes shorter, the dynamic load factor becomes less than two, decreasing as the decay time decreases. In the limit of very short pressure pulses, the loading is in the impulsive regime and the peak elastic deformation is proportional to the impulse. Between these two extremes, in the mixed regime, the peak elastic deformation will depend on both the impulse and peak pressure.

Evaluation of the experimentally determined pressures from the empty thick-walled tube provides DLFs between 1.2 and 2.6 for mixture A. The evaluation of the thick-walled tube with concentric annular gap provides DLFs between 0.7 and 1.8 for experimentally determined pressures values. The dynamic load factors of the annulus configuration are less than the DLFs for the empty tube. One reason is that the gas volume for the annular gap is only 7.5% of the empty tube so that the total energy released in the combustion event is much smaller in the annulus than in the empty tube. Another reason is that DDT was initiated promptly for the annulus configuration, so the detonation was approximately an ideal CJ wave when it propagated to the tube end, while for the empty tube; the detonation wave was highly overdriven due to the DDT event.

3013 Container Testing

As a confirmation of the applicability of the test results, actual 3013 containers were instrumented with strain-gages and fitted with pressure ports to measure structural loading and response to deliberate ignition of the explosive mixtures. Figure 11 provides a photo of the modified 3013 container and a drawing of the test setup. Filling the 3013 container, which is the outermost container and has the largest volume, with the various explosive mixtures was considered to provide the worst case structural loading for the storage system because it maximizes the energy content within the system. The presence of the inner containers, not included in this test, not only reduces the gas volume but also acts as energy absorbing media, thus reducing the energy absorbed by the outer container. These observations demonstrate that the assumption of filling the empty 3013 container with the explosive mixture as the worst case condition for evaluating loss-of-containment for the system is justified.



spark plug hole

3013 outer can

PCB adaptor

PCB adaptor



Figure 11 (a) Modified 3013 outer can. (b) Drawing showing modified 3013 outer can with instrumentation locations. 1-3013 outer can, 2-welded flange, 3-spark/glow plug, 4-pressure transducer adapters, 5-strain gauges, 6-thermocouple, 7-static pressure gauge, 8 and 9-gas fill/circulation lines.

Figure 12 shows the recorded peak pressures on pressure transducers P1-P5, and peak strains on S1-S9 for all the shots and mixtures.



Figure 12 Comparison of peak pressures and strains for three mixtures of the 3013 empty can tests.

For the empty 3013 outer can configuration, the DDT transition was observed at an initial pressure of 2.6- 2.7 bar for mixture A. This is essentially the same threshold as observed for the empty thick-walled fixture (Figure 6a)). The maximum peak strain was usually observed near the middle of the can on either S1 or S2 instead of close to the reflecting end as observed for the thick-walled fixture (Figure 6b). Peak strain increases with increasing initial pressure, and the overall trend is linear with sharp increases in the vicinity of the DDT threshold. Below the threshold at initial pressure of 2.6 bar, the peak strain was on the order of 700 µstrain. Above the threshold at an initial pressure of 2.7 bar, the peak strain was on the order of 1800 µstrain, very close to the convention for the onset of plastic behavior (2000 µstrain). For mixtures B and C, no DDT transition was observed for initial pressures up to 3.5 bar, which is consistent with the findings with the thick-walled tube.

The DLF in terms of the experimental measured pressures for the 3013 container tests ranged between 0.4 and 1.2 (~1.2 and 2.6 for the empty thick-walled tube configuration). The values obtained indicate mixed mode loading between the impulsive and sudden regimes. The slightly higher values measured for the thick-walled tube configuration are most likely due to differences in the structural response associated with detonation loads.

In Figure 13, the measured strains are compared with estimated strains based on P_{CJ} with dynamic load factors of 1 (static loading), 2 (sudden loading) and 5 (reflected detonation). For the empty 3013 container within the DDT range (initial pressure > 2.6 bar), the maximum measured strains are all larger than $\mathcal{E}_{CJ,\Phi=2}$, which is consistent with the results from the thick-walled tube. This is because DDT occurred close to the tube end, producing much higher strains than the case where detonation was initiated promptly.



Figure 13 Comparison between the measured strains and the estimated strains (\mathcal{E}_{CJ}) based on P_{CJ} and Φ =1, 2, and 5 for mixture A of the 3013 empty cans tests.

Discussion

For the 3013 storage containment system, deflagration-to-detonation transition is possible both within the annulus between the inner and outer containers for all mixtures tested as demonstrated by the testing of the planar fixture and the thick-walled cylinder tests with annular gaps. Deflagration-to-detonation transition was also observed in the empty thick-walled cylinder tests and the actual 3013 container tests (without an inner container) at sufficiently high initial pressure with stoichiometric hydrogen-oxygen mixtures.

For the three mixtures tested, the peak hoop strains measured in the outer 3013 container are slightly less than the 0.2% strain conventionally used to determine the onset of plastic deformation. No structural failure or measurable deformation was found in the 3013 outer containers that were tested. Based on the results of these tests, it can be concluded that DDT of a stoichiometric hydrogen-oxygen mixture (and mixtures diluted with nitrogen and helium) within the 3013 nested containment system does not pose a threat to structural integrity of the outer container at initial pressures up to 3.5 bar and temperatures up to 150 °C.

The inner or convenience containers were not tested. Based on these test results and analytical studies (Liang and Shepherd 2007), the DDT threshold initial pressures are expected to be lower for small diameter containers and containers filled with granular material. Because peak pressures are proportional to initial pressures, the peak DDT pressures measured in the 3013 outer containers will bound the peak DDT pressures that will occur in the inner and convenience containers. If an explosion were to occur in the inner or convenience containers the peak strains and deformations will be higher for the inner and convenience containers than for the outer container because the outer container is more robust structurally than the inner and convenience containers.

Conclusion

The 3013 outer container is the credited safety pressure boundary for the nested 3013 storage canister system. The test results show that integrity of the 3013 container system will maintain its structural integrity following the postulated explosion accident.

Acknowledgements

Funding for this work was provided by the Surveillance and Monitoring Program, US Department of Energy Office of Environmental Management. This work was conducted at California Institute of Technology, *Los* Alamos National Laboratory operated by Los Alamos National Security, LLC under contract DE-AC52-06NA25396, and at the Savannah River National Laboratory operated by Savannah River Nuclear Solutions for US Department of Energy under contract DE-AC09-08SR22470.

References

Z. Liang and J.E. Shepherd, "Explosion Testing of Nested Can Containment System" in 3 parts, Report No. FM2007.001-.003, Explosion Dynamics Laboratory, California Institute of Technology, May 9, 2007.

J. Biggs. Introduction to structural dynamics. McGraw-Hill, Inc., 1964. ISBN 07-005255-

M. Paz and W. Leigh. Structural Dynamics. Springer, fifth edition, 2004.

W.C. Reynolds. The element potential method for chemical equilibrium analysis: implementation in the interactive program STANJAN. Technical report, Mechanical Engineering Department, Stanford University, 1986.