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ANALYTICAL PREDICTION AND VERIFICATION OF HYDROGEN-AIR DEFLAGRATION PRESSURES RESULTING FROM TRANSURANIC WASTE RADIOLYSIS INSIDE OF SEALED CONTAINERS

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ABSTRACT

Maintaining flammable gas concentrations below flammable limits in the layers of confinement of transuranic (TRU) waste is a limiting factor in the efficient Type B shipment of nuclear materials. Traditionally, flammable gas limits are maintained by limiting hydrogen generation due to radiolysis in the waste matrix, restricting the quantity of flammable volatile organic compounds in the container headspace, and implementing prohibitions on aerosol cans and large sealed containers. These restrictions have been driven by the requirements of 10CFR71.43(d) to ensure that no significant chemical reaction could occur among the packaging contents.

The response of the Type B package containment boundary to pressures generated as a result of a deflagration event is of particular interest when evaluating alternatives to the restrictive payload controls traditionally implemented. An analytical method that utilizes the Cheetah™ Thermo-chemical Kinetics code, developed by Lawrence Livermore National Laboratory, has been used to determine the deflagration pressures resulting from a stoichiometric hydrogen-air concentration inside of a sealed container under adiabatic constant volume assumptions. The adiabatic constant volume deflagration pressure is then adjusted to account for the void volume outside of the sealed container to determine the pressures exerted on the packaging containment boundary.

A series of full-scale stoichiometric hydrogen-air deflagration tests have been performed to validate the analytical deflagration pressure prediction methodology. It is demonstrated that the analytical method can be utilized to conservatively predict the pressure that a sealed container of a given size and pressure capacity exerts on the packaging containment boundary when undergoing a stoichiometric hydrogen-air deflagration.

INTRODUCTION

Layers of confinement in the TRU waste payloads can potentially contain flammable concentrations of fuel and oxidizer. The fuel is hydrogen generated due to radiolysis in the payload, the content and propellant in aerosol cans, and/or volatile organic compounds released from the waste matrix. The oxidizer is the oxygen present in atmospheric air inside the layer of confinement prior to closure. In the presence of an ignition source, a flammable gas mixture can potentially deflagrate within the sealed container causing pressure build-up due to flame propagation and burning in the confined volume.

Confinement layers in TRU waste payloads can broadly be categorized as follows:

- **Sealed Container** – Any waste packaging boundary greater than 4 liters in size that is assumed to prohibit the release of gas across the boundary. A waste packaging component meeting this definition does not have a known release rate of hydrogen gas out of its confined space. Examples of sealed containers are rigid unfiltered containers with fully-welded or gasketed lid closures.
- **Unsealed Layer of Confinement** – Any waste packaging boundary that restricts, but does not prohibit, the release of gas across the boundary. A waste packaging component meeting this definition has a known release rate of hydrogen gas out of its confined space. Examples of unsealed layers of confinement are twist-and-tape plastic bags, heat-sealed plastic bags, filtered plastic bags, and metal containers or drums

fitted with filters. Waste packaging materials that allow for the free release of gas (e.g., punctured plastic bags, bags open at the end, pieces of plastic sheeting wrapped around the waste for handling, and metal containers with lid closures that allow free gas release) do not meet this definition and are simply considered to be part of the waste.

For TRU waste that is comprised of a mixture of sealed containers and unsealed layers of confinement, two different approaches can be utilized to control and characterize the potential for a flammable mixture deflagration within the waste, respectively. Application of a controlled payload evacuation process followed by backfilling with an inert gas is the process of choice for eliminating the potential for a deflagration inside unsealed layers of confinement. The objective of the evacuation/backfill process is to reduce the concentration of oxygen (i.e., a potential oxidizer) within any unsealed layers of confinement such that no flammable mixture and no possibility of a deflagration event exists within these void spaces during the shipping period. Due to the lack of a known gas release rate across the boundary of a sealed container, the evacuation and backfill process can not be relied upon to remove the oxidizer to below flammable levels and prohibit a deflagration inside of a sealed container. As such, a bounding approximation to the pressures exerted by a sealed container deflagration must be established and controlled to less than the maximum normal operating pressure (MNOP) of the Type B package containment vessel in order to ensure the safe transport of potentially flammable mixtures of gases in Type B packages.

ADIABATIC CONSTANT VOLUME DEFLAGRATION PRESSURE MODEL

This section presents a model that conservatively estimates the pressure build-up in a Type B package containment vessel (CV) resulting from a stoichiometric constant volume deflagration inside of a sealed container as a function of the initial size and pressure of the sealed container.

Fuel and Fuel/Oxidizer Ratio

The adiabatic constant volume deflagration pressure model utilizes hydrogen as the fuel at a stoichiometric fuel/oxidizer ratio. Hydrogen is chosen as the fuel for the model due to its predominance as a flammable gas in TRU waste and its high heat of combustion in comparison with other flammable gases such as ethane, propane, and ethylene. Additionally, hydrogen represents the most energetic flammable gas for a deflagration in a sealed container due to its very high laminar burn velocity in comparison with other flammable gases such as ethane, propane, ethylene, and acetylene.[1]

A stoichiometric mixture of fuel (hydrogen) and oxidizer (oxygen in air) is chosen as the mixture that produces the highest adiabatic constant volume combustion pressure and temperature attributed to complete combustion of the reactants. The stoichiometric combustion reaction of hydrogen with air is presented by the following chemical equation:

$$2H_2 + O_2 + 3.76N_2 \rightarrow 2H_2O + 3.76N_2$$

Therefore, the volume percent of hydrogen in air (21% O_2, 79% N_2) required to produce a stoichiometric hydrogen-and-air mixture is as follows:

$$\%H_2 = \frac{\text{mol}(H_2)}{\text{mol}(\text{Air}) + \text{mol}(H_2)} \times 100 = \frac{2}{(1+3.76)+2} \times 100 = 29.58\%$$

The volumetric air-to-hydrogen ratio for a stoichiometric mixture is similarly calculated as follows:

$$\frac{V_{\text{Air}}}{V_{H_2}} = \frac{\text{mol}(\text{Air})}{\text{mol}(H_2)} = \frac{(1+3.76)}{2} = 2.38$$

Cheetah™ Adiabatic Constant Volume Deflagration

Figure 1 (from Shaw[1]) gives the adiabatic constant volume hydrogen deflagration (combustion) pressures and temperatures as a function of equivalence ratio (fuel concentration / stoichiometric fuel concentration). The combustion temperatures and pressures were calculated by Cheetah™ 4.0, a thermochemical-kinetics code developed by Lawrence Livermore National Laboratory.[2] The results are presented for initial conditions at 1 atm and 293 K. The ordinate of the figure for pressure can alternatively be interpreted as a pressure factor and multiplied by any initial absolute pressure to determine the resulting deflagration absolute pressure.

As seen in Figure 1, the adiabatic constant volume deflagration pressure and temperature is maximized when the equivalence ratio is unity such that the hydrogen concentration in air is 29.58%. Correspondingly, the pressure increase factor, P_{factor}, for an adiabatic constant volume hydrogen deflagration at stoichiometric conditions is 8.18.

The Cheetah™ results are valid for and can be applied to any fixed volume since the calculations are based on a given fuel mixture density.

Void Volume Scaling

Under the assumption that a sealed container undergoes a stoichiometric deflagration and releases the combustion gases into the void space of an internal payload container, which in turn releases the combustion gases into the void space in the package containment vessel, Boyle's Law can be used to conservatively calculate the resulting pressure increase experienced by the containment vessel as a result of the deflagration. This approach attributes the pressure to heat-up and expansion of combustion gases as predicted by the thermochemical-kinetics code with pressure reductions due to a progressively increasing void space (i.e., sealed container to payload container to containment vessel) and neglects any overpressure due to the propagating flame front. Due to the length scales, modest ignition energy potential in the payload, and geometric aspect ratio of the sealed container(s), payload container, and containment vessel, the deflagration assumption is valid. Neglecting the flame front overpressure is appropriate when combined with other model conservatisms since a spherical flame front must travel at exceedingly high effective burning velocities (over 20 meters per second or about 50 times the normal burning velocity of most hydrocarbons) before damaging blast waves (overpressures >0.3 atm) can be generated by a deflagration.[3]

The gauge pressure generated inside a sealed container, P_{sc_defl}, from an adiabatic constant volume deflagration under stoichiometric conditions is given as a function of the initial gauge pressure inside the sealed container, P_{sc_init}, and atmospheric pressure, P_{atm}, as follows:

$$P_{sc\_defl} = [P_{factor} \times (P_{atm} + P_{sc\_init})] - P_{atm} \qquad [1]$$

If the sealed container were to breach during the deflagration and release gases into the payload container, the gauge pressure generated inside the payload container, P_{pc_defl}, is proportional to the increase in volume available for gas expansion per Boyle's Law. For a given void volume in the sealed container, V_{sc_void}, and void volume in the payload container, V_{pc_void}, the final gauge pressure in the payload container is given as follows:

$$\begin{aligned} P_{pc\_defl} &= \frac{([P_{atm} + P_{sc\_defl}] \times V_{sc\_void}) + (P_{atm} \times V_{pc\_void})}{(V_{sc\_void} + V_{pc\_void})} - P_{atm} \\ &= P_{sc\_defl} \times \frac{V_{sc\_void}}{(V_{sc\_void} + V_{pc\_void})} \end{aligned} \qquad [2]$$

Correspondingly, if the payload container were to breach and release gases into the containment vessel, the gauge pressure generated inside the containment vessel, P_{cv_defl}, is additionally given as a function of the void volume in the containment vessel, V_{cv_void}, as follows:

$$\begin{aligned} P_{cv\_defl} &= \frac{([P_{atm} + P_{pc\_defl}] \times [V_{sc\_void} + V_{pc\_void}]) + (P_{atm} \times V_{cv\_void})}{(V_{sc\_void} + V_{pc\_void} + V_{cv\_void})} - P_{atm} \\ &= P_{sc\_defl} \times \frac{V_{sc\_void}}{(V_{sc\_void} + V_{pc\_void} + V_{cv\_void})} \end{aligned} \qquad [3]$$

The above approach conservatively over-predicts the maximum pressure due to heat-up and expansion of combustion gases from a sealed container deflagration within a payload container inside a Type B package. The constant volume pressure factor is based on an adiabatic assumption that neglects the energy absorbed in breaching the sealed container and payload container and any heat losses into the payload, payload container, or packaging. The void volumes referenced above are shown schematically in Figure 2.

Application to Sealed Container Inventory

For a given package containment vessel, payload container, and sealed container where the void volumes are known, the selection of a bounding initial gauge pressure inside the sealed container is the only additional variable that needs to be defined. The pressure associated with a stoichiometric deflagration of hydrogen and air inside a sealed container is limited to the pressure increase due to hydrogen generation required to achieve stoichiometric conditions that is above the one atmosphere of air that could be present in the sealed container when sealed. For conservatism, the above methodology utilizes a bounding initial gauge pressure inside the sealed container equal to the burst pressure of the sealed container as determined by test. The burst pressure is defined as the either the maximum pressure associated with burst (gross structural failure) of the container or a pressure that is limited by equilibrium between the internal gas generation rate and external leakage rate from the container. The minimum input flow rate utilized in sealed container hydrostatic testing is conservatively selected to ensure that the test conditions for input flow rate exceed the potential gas generation rate in the sealed container.

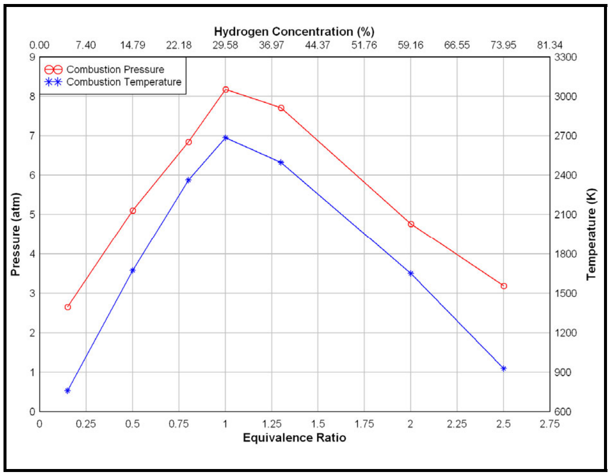

Figure 1. Cheetah-Computed Deflagration Pressure and Temperature

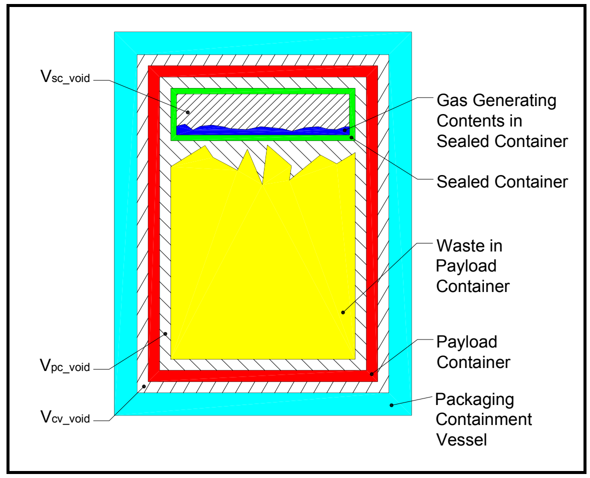

Figure 2. Void Volume Schematic

DEFLAGRATION PRESSURE TESTING AND PRESSURE MODEL VALIDATION

Stoichiometric hydrogen deflagration tests were performed utilizing a large surrogate sealed container (SSC) designed to initially contain the pressurized hydrogen/air mixture, a prototypic payload container (SLB2), a rigid dunnage assembly (SLB2 dunnage) designed for testing to consume void space inside the SLB2, and a mock-up of the TRUPACT-III containment vessel (Mock CV) designed as a vessel to contain and facilitate measurement of the deflagration pressure. The testing was performed at the Energetic Materials Research and Testing Center (EMRTC) at New Mexico Tech University in Socorro, New Mexico.[4,5] The test articles were sized utilizing the model developed above to theoretically produce a pressure in the Mock CV equal to the MNOP of the TRUPACT-III CV. Two variations on the test configuration were implemented, one with filter vents installed and one with the filter ports open in the SLB2 to determine whether the measured Mock CV pressures were affected by the rate of combustion gas throttling through the vent port/filter openings. The objectives of the testing were to 1) compare the pressure measured in the Mock CV void space to validate the pressure predicted by the adiabatic constant volume deflagration model and 2) observe the structural response of the SLB2 and Mock CV to a deflagration test environment that was more severe than what would potentially exist in the actual TRUPACT-III payload.

Test Set-up

As illustrated in Figure 3, the deflagration test assembly consisted of an SSC placed inside and bolted to the SLB2 via a gasketed instrumentation flange, SLB2 dunnage placed inside of the SLB2, and the SLB2 placed inside and bolted to the Mock CV via a gasketed instrumentation flange. Eight (8) piezoelectric dynamic pressure transducers were installed in the test assembly; five located in the Mock CV coupling ports (labeled P1 thru P5) with three located in the Mock CV, SLB2, and SSC instrumentation flanges (labeled P6 thru P8, respectively) as shown in Figure 4. The piezoelectric dynamic pressure transducers (PCB Piezotronics Model No. 102A05) are ICP® (Integrated Circuit Piezoelectric) quartz crystal voltage-mode type sensors with built-in microelectronic amplifiers that convert high-impedance charge into a low-impedance voltage output. The outputs of all dynamic pressure transducers were attached via coaxial cable to a digital data acquisition system (PCB Piezotronics Model No. 481A) capable of capturing data at a rate of 125,000 samples per second. Also located in the SSC instrumentation flange was an electric match port where the electric match leads were passed through the port, leaving the head of the match extending into the center of the SSC. One of the ports in the SSC instrumentation flange was utilized to connect a static pressure transducer that incorporated tubing and valving to isolate the pressure transducer to measure the pressure inside the SSC and open the system to atmosphere to allow ambient pressure measurements. The last port in the SSC instrumentation flange was utilized to accommodate the hydrogen/air fill hose, which was valved and connected to dry air and hydrogen gas sources. The assembled instrumentation flange is shown in Figure 5, and the overall plumbing arrangement is shown in Figure 6. Additionally, a thermocouple was utilized both in free air and attached to the flange end sidewall of the Mock CV for measuring ambient and Mock CV wall temperatures.

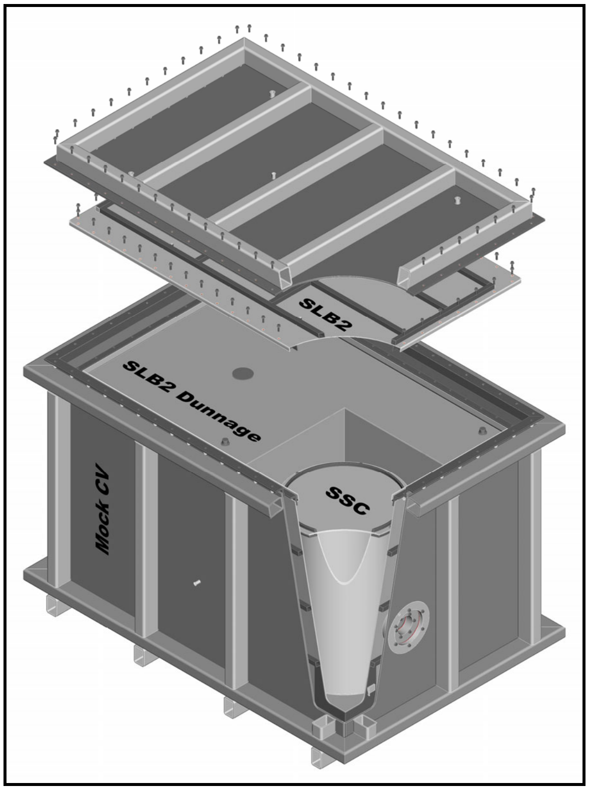

Figure 3. Deflagration Test Assembly

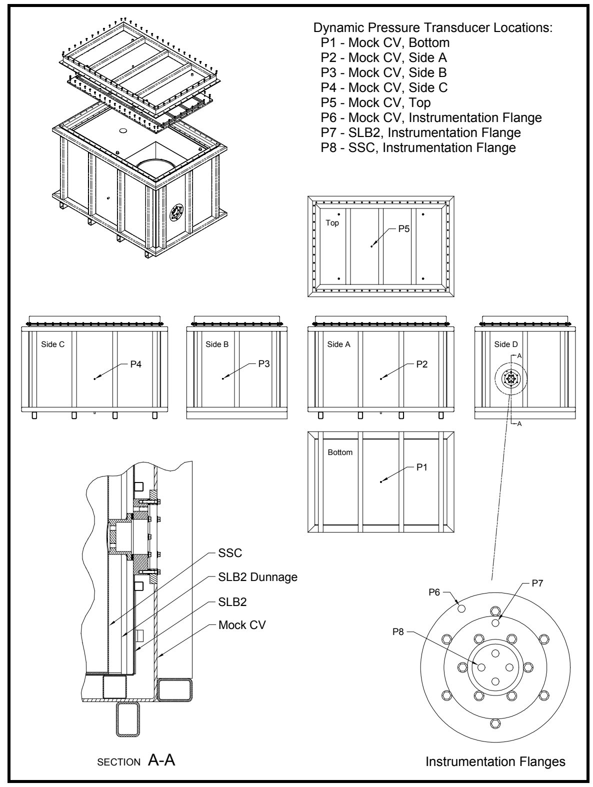

Figure 4. Transducer Map

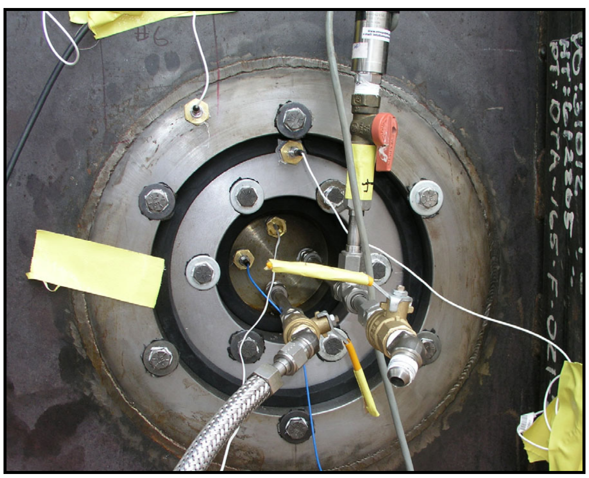

Figure 5. Instrumentation Flange

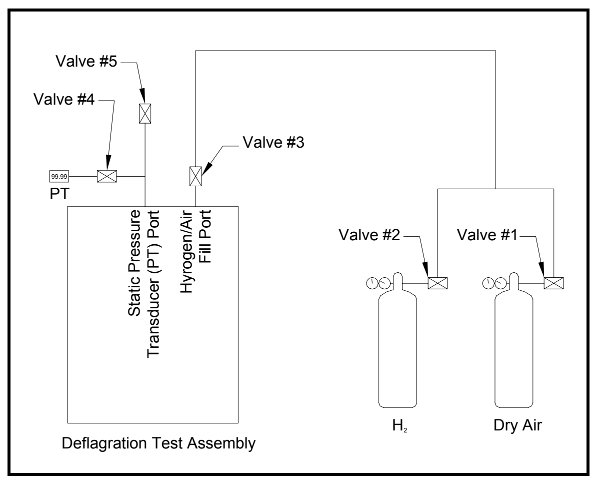

Figure 6. Plumbing Schematic

Technical Basis

The test articles were sized utilizing the deflagration pressure model presented above to theoretically produce a pressure inside the Mock CV of 25 psig. The gauge pressure of the SSC required to achieve a stoichiometric mixture, by adding hydrogen to the air initially present in the SSC at ambient conditions, is given as follows:

$$P_{ssc\_init} = P_{atm} \times \left(1 + \frac{1}{V_{Air} / V_{H_2}}\right) - P_{atm} = 14.7 \times \left(1 + \frac{1}{2.38}\right) - 14.7 = 6.17 \text{ psig}$$

The SSC size was determined by satisfying equation [1], equation [2], and equation [3] using the stoichiometric pressure increase factor of 8.18 to theoretically produce a Mock CV pressure equal to 25 psig. Overall, the technical basis for the confirmatory deflagration testing is consistent with the bounding conservatism in the adiabatic constant volume deflagration pressure model. The test was designed to maximize the deflagration pressure increase factor by igniting a stoichiometric hydrogen/air mixture. Hydrogen was chosen as the test flammable gas because of its high heat of combustion and its high laminar burn velocity. The test was designed to absorb minimal energy in releasing the combustion gases from the sealed container through the utilization of an engineered lid release mechanism. It utilized rigid dunnage to consume 75% of the void space in the SLB2 surrounding the SSC and, by avoiding significant contents crushing, minimized the void space and maximized the pressures seen by the SLB2 and Mock CV. Also, the Mock CV was utilized to replicate the TRUPACT-III CV void volume and resist significant deformation to provide an equivalent void space for measuring the deflagration pressures. Finally, the prototypic SLB2 was evaluated utilizing two configurations of the filter ports (filter vents installed, filter ports open) to evaluate the effects of combustion gas throttling through the most likely paths of leakage from the SLB2 into the Mock CV.

Test Results and Conclusions

In addition to the measurement and recording of the deflagration pressures, each test article comprising the deflagration test assembly was visually inspected to determine the response of each to the deflagration event. For both tests, the Mock CV experienced no obvious structural damage and passed the post-deflagration pressure decay test to demonstrate the closure integrity. The SLB2 payload container experienced significant permanent deformation of all sides with damage of the external bumpers resulting from dynamic impact with the Mock CV. The SSC lid bolts separated as designed and released the SSC lid into the SLB2 lid with only superficial interaction marks indicating the impact. A summary of the structural damage to the SSC and SLB2 is provided in Figures 7 and 8, respectively.

For the "ports-filtered" test and inspection of Figure 9, the average pressure exerted on the Mock CV (reported by transducers P1, P3, and P6) from 25 to 100 msec ranges from 6.6 psig to 12.7 psig with a sustained average over the 75 msec time duration of 10.4 psig. Due to the adiabatic assumption in the analytical model, the analytical predicted deflagration pressure (22 psig when accounting for test condition variations in void volume, atmospheric pressure, etc.) is conservatively overestimating the average pressures experienced by the Mock CV during the deflagration event.

For the "ports-open" test and inspection of Figure 10, the average pressure exerted on the Mock CV (reported by transducers P1, P3, and P6) from 25 to 100 msec ranges from 4.8 psig to 11.7 psig with a sustained average over the 75 msec time duration of 10.0 psig. Due to the adiabatic assumption in the analytical model, the analytical predicted deflagration pressure (20 psig when accounting for test condition variations in void volume, atmospheric pressure, etc.) is conservatively overestimating the average pressures experienced by the Mock CV during the deflagration event.

The tracking of the pressure results between the two tests suggests that the primary factor affecting Mock CV pressure is the void volume that the combustion gases are available to expand into and not primarily the mechanism for release of those gases from the SLB2. Contrastingly, when comparing the pressure traces for both tests in the SSC (P8) and SLB2 (P7), it is clear that the pressures seen by the SLB2 are affected by the flow of combustion gases through the filter ports. The more restrictive flow out of the SLB2 in test #1 results in higher peak pressures in the SLB2 when compared to test #2. In both cases, the magnitude of pressures inside the SLB2 causes significant "ballooning" of the SLB2 and structural interaction between the SLB2 and the Mock CV, as ascertained from the damage assessment of the SLB2 bumpers. Therefore, the test data demonstrates that the analytical model is appropriately conservative, even when compared to a test case where the combustion gases are allowed to flow freely through open SLB2 filter ports into the Mock CV.

Figure 7. Post-Test SSC

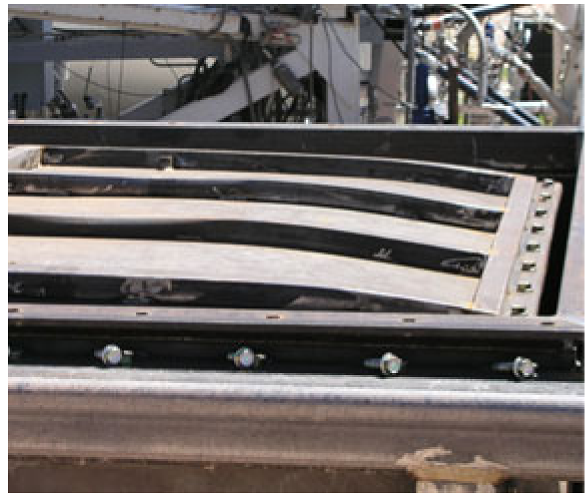
Figure 8. Post-Test SLB2

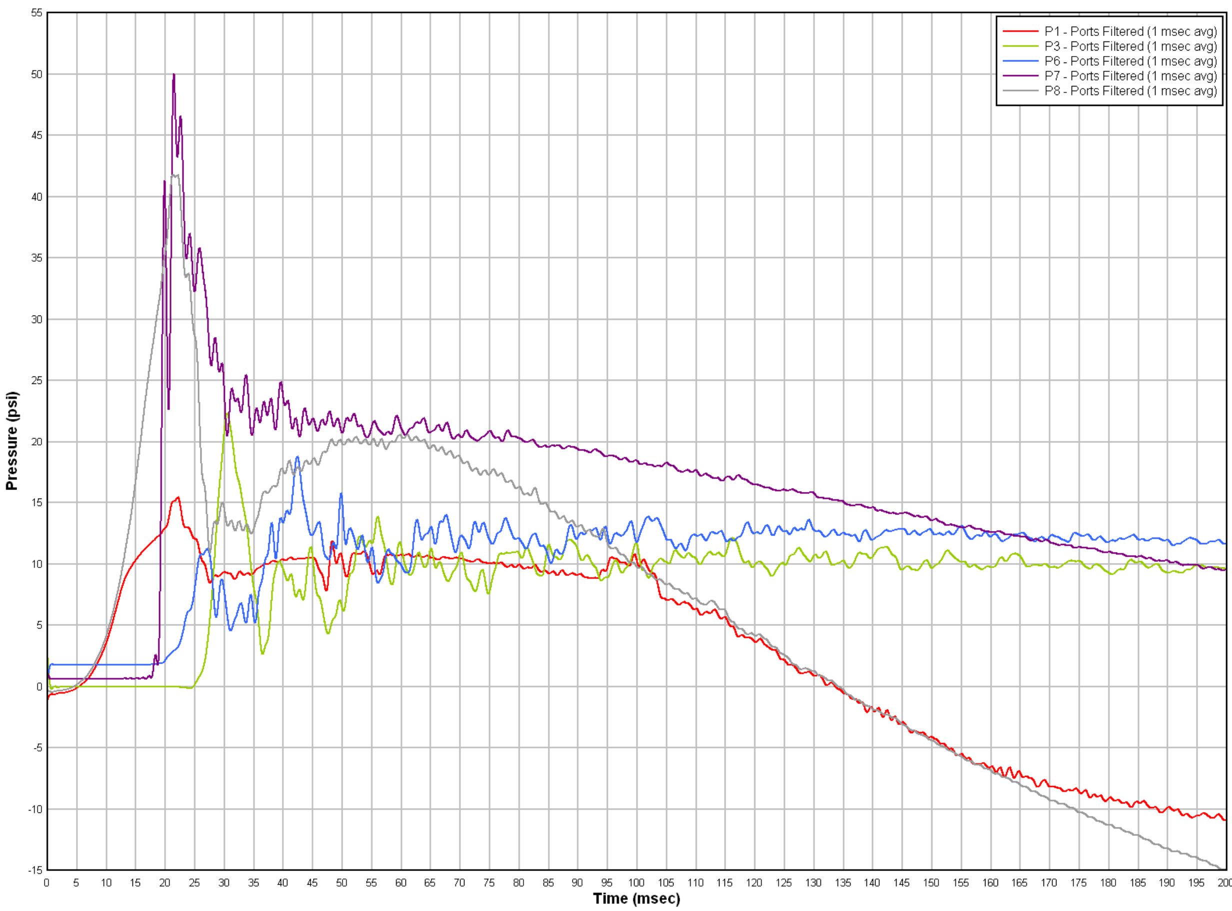

Figure 9. "Ports Filtered" Pressure Transducer Data

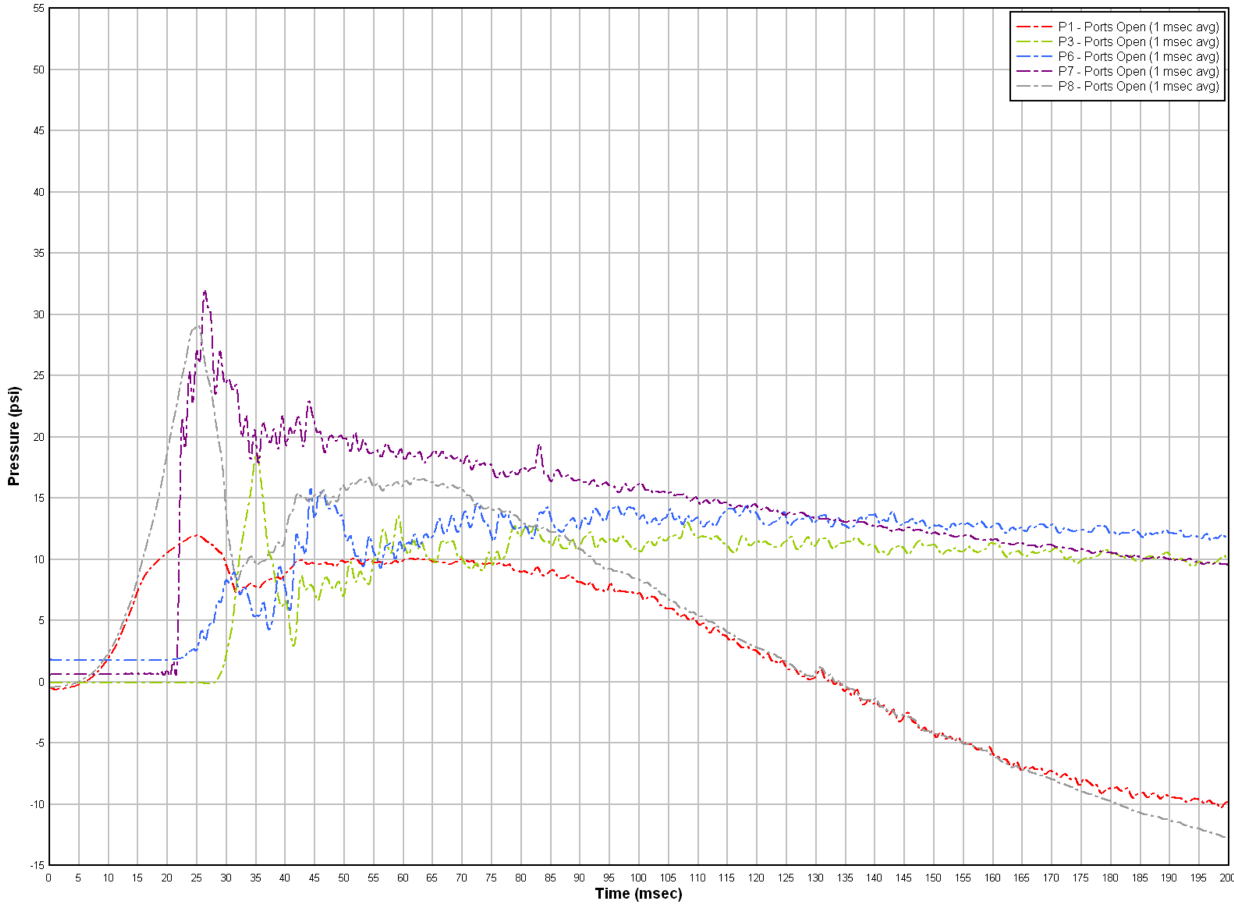

Figure 10. "Ports Open" Pressure Transducer Data

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