

April 15, 2013

Ms. Huda Akhavannik – Project Manager
Office of Nuclear Material Safety and Safeguards
Mail Stop: EBB-3D-02M
United States Nuclear Regulatory Commission
Executive Boulevard Building
6003 Executive Boulevard
Rockville, Maryland 20852

RE: Revision 1 – Evaluation of Thermal Degradation of Packaging Material in Versa-Pac, Request to Amend Certificate of Compliance No. 9342 for the Model No. Versa-Pac Package and Drawing Corrections to the SAR under Docket No. 71-9342

Dear Ms. Akhavannik,

| Certificate Number | Model Number |
|--------------------|----------------|
| USA/9342/AF | VP-55 & VP-110 |

Please find enclosed a revised copy of the DAHER-TLI Engineering Services report Titled: Evaluation of Thermal Degradation of Packaging Material in Versa-Pac, CN-13002-301 Revision 1 which provides the corrections in the room temperature used in the calculations in support of our request for amendment to the Certificate of Compliance Revision 6 and Safety Analysis Report Revision 6.

The corrected engineering report calculations do not change the final conclusions previously drawn from the original report and are respectfully submitted for your continued review and approval.

If you or the staff have any questions, or need any additional information, please let me know.

Respectfully,

William M. (Mike) Arnold

Mike

William M. (Mike) Arnold
Phone: 423-646-1864
E-Mail: CenturyIndWMA@aol.com

PROJECT NAME:

13002 - Versa-Pac

CLIENT:


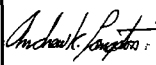
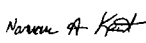
Century Industries

CALCULATION TITLE:

Evaluation of Thermal Degradation of Packing Material in Versa-Pac

PROBLEM STATEMENT OR OBJECTIVE OF THE CALCULATION:

To demonstrate that the integrity of the Versa-Pac inner containment will not be exceeded due to heat and gas produced by the thermal degradation of packing materials.

| Revision | Affected Pages | Revision Description | Name of Preparer(s) & Checker(s) | Engineering Manager Approval/Date |
|----------|-------------------|--|--|---|
| 0 | 1 - 26 | Initial issue | Peter J. Vescovi Andrew K. Langston | Norman A. Kent |
| 1 | 6,13,18,20, 21-26 | Correct error in calculation. Incorrect temperature used for room temperature. Assumed temperature for auto-ignition changed to calculated wall temperature for the inner container. | Peter J. Vescovi  Digitally signed by Peter Vescovi DN: cn=Peter Vescovi, o=TLI Engineering Services, ou=Chief Engineer, email=pvescovi@tlusa.com, c=US Date: 2013.04.12 13:55:52 -05'00' Andrew K. Langston  Digitally signed by Andy Langston DN: cn=Andy Langston, o=TLI Engineering Services, email=alangston@tlusa.com, c=US Date: 2013.04.12 13:57:23 -05'00' | Norman A. Kent  Digitally signed by Norman Kent DN: cn=Norman Kent, o=TLI Engineering Services, ou=Director, email=nkent@tlusa.com, c=US Date: 2013.04.12 13:57:02 -05'00' |

INDEPENDENT DESIGN VERIFICATION CHECK SHEET

Calculation Note Number: CN-13002-301
Scope of Analysis File: Evaluation of Thermal Degradation of Packing Material in Versa-Pac

Design Review Methodology:

Check of Calculations: ☒
 Alternate Calculation: ☐
 Other (explain): ☐

Confirm that the Calculation Package Reviewed Includes:

1....Statement of Purpose ☒
 2....Defined Method of Analysis ☒
 3....Listing of Assumptions ☒
 4....Detailed Analysis Record ☒
 5....Statement of Conclusions / Recommendations (if applicable) ☒
 6....References ☒

| Step | Activities | Verification | | | Comments |
|------|---|-------------------------------------|--------------------------|-------------------------------------|----------|
| | | Yes | No | N/A | |
| 1 | For the scope of the defined analysis: A. Are the required data input complete? • Material Properties • Geometry (Drawing Reference) • Loading Source Term If a supporting analysis is required to define the load state, has it been defined? B. Are Boundary conditions acceptable? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 2 | Is the method of analysis adequate for the defined scope? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 3 | Is the worst-case loading/configuration documented? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 4 | Are the acceptance criteria defined and complete? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 5 | Has all concurrent loading been considered? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 6 | Are analyses consistent with previous work for method and approach? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 7 | Are the records for input and output complete? | <input type="checkbox"/> | <input type="checkbox"/> | <input checked="" type="checkbox"/> | |
| 8 | Has the computer output been verified? | <input type="checkbox"/> | <input type="checkbox"/> | <input checked="" type="checkbox"/> | |
| 9 | Is traceability to verified software complete? | <input type="checkbox"/> | <input type="checkbox"/> | <input checked="" type="checkbox"/> | |
| 10 | Is the statement of conclusions and recommendations complete and acceptable for the project and objectives of the defined purpose? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 11 | Are references complete? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 12 | Are results reasonable for purpose of calculation? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 13 | Have all assumptions been justified and confirmed? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |
| 14 | Has the cumulative effect of specified dimension tolerances on the design been addressed? | <input checked="" type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> | |



Digitally signed by Andy Langston
 DN: cn=Andy Langston, o=TLI Engineering Services, email=alangston@tlusa.com, c=US
 Date: 2013.04.12 13:57:44 -05'00'

Reviewer (Electronic Signature and Date)

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NOTICE

Transport Logistics International, Inc. (TLI) is a majority owned subsidiary of Nuclear Cargo and Services, GMBH (NCS) and is part of the DAHER group of companies.

1. INTRODUCTION

Fissile material packages are typically evaluated using water as the moderating material. The implied assumption is that there are no other moderating materials mixed with the contents during routine, normal, and accident transport conditions that are more effective than water. Limits may be imposed on the quantity of moderating material, other than water, that are based on assumptions made for the criticality evaluation. These limits may be specified in the approval certificate, either as a maximum limit on the hydrogen density of the moderator, or as a limit on the quantity of moderating material.

Versa-Pac Background

The Versa-Pac is evaluated assuming optimum moderation with high-density polyethylene (HDPE) plastic (density = 0.98 g/cc) rather than water. The Versa-Pac approval certificate [1] USA/9342/AF-96, initially specified the limit on polyethylene as:

5.(b)(1) Type and Form of Material – Revisions 0 – 3

Contents may be pre-packaged in polytetrafluoroethylene, aluminum, and carbon steel per Table No. 1-4 of the application.

Payload materials shall have an auto-ignition temperature and melting point greater than 600°F. Materials with a hydrogen density greater than 0.141g/cm³ are not authorized.

The Versa-Pac certificate of compliance (CoC) supports packaging applications containing both carbon-based (e.g., graphite, paraffin, and polyethylene) and hydrogen-based materials (e.g., water paraffin, and polyethylene) [2]

The density of hydrogen in the HDPE (0.141 g/cm³) is higher than the density of hydrogen in water (0.112 g/cm³ at 25°C). The limits on the auto-ignition temperature and melting point are imposed to prevent combustion of materials that could result in higher temperatures and pressures than evaluated for the BU-D. The maximum temperature expected during the accident condition fire is 360°C (680°F).

A subsequent need to use additional packing materials led to specifying other materials in the contents as:

5.(b)(1) Type and Form of Material – Revisions 4 and 5

Contents may be pre-packaged in polytetrafluoroethylene, aluminum, and carbon steel per Table No.1-4 of the application. Aluminum Trihydrate and Sodium Borate (Borax, fused) are also authorized as packing materials.

Payload materials shall have an auto-ignition temperature and melting point greater than 600°F. Materials with a hydrogen density greater than 0.141 g/cm³ are not authorized.

Further consideration of the materials used to package and label the contents led to a restriction on the these materials as:

5.(b)(1) Type and Form of Material – Revisions 6

Contents may be pre-packaged in polyethylene, polytetrafluoroethylene, aluminum, and carbon steel per Table No.1-4 of the application. Aluminum Trihydrate, Sodium Borate (Borax, fused), perlite, paper labels, plastic tape, plastic bags, plastic bottles and desiccant such as "Quik-Solid®" are also authorized as packing materials. Materials with a hydrogen density greater than 0.141 g/cm³ are not authorized.

The hydrogenous packing material load in the form of paper and plastics is limited to a total of 200 grams per package.

Radioactive contents shall have an auto-ignition temperature and melting point greater than 600°F.

The limit of 200 grams per package is an arbitrary quantity chosen to limit the temperature increase and subsequent pressure increase inside the inner containment. There is no quantitative evaluation to demonstrate that combustion of 200 grams of paper or plastic would increase the temperature enough to exceed the design pressure of 15 psig.

Previous Amendment Request

During the review of a previous amendment request, the NRC made the following conclusions [3] with regard to auto-ignition and quantity of hydrogenous material:

Auto-Ignition

Maximum calculated weighted payload temperature of 360.4°F (at 22 minutes into the fire) is well below the auto-ignition temperatures of 424~474°F for paper, 660-986°F for plastics

Package vessel provides a barrier to exclude auto-ignition (600°F) when the maximum local temperature may be around 552°F under HAC fire.

Percentage of hydrogenous material load to the allowable payload

Plastics with an auto-ignition temperature greater than 660°F will not auto-ignite under a weighted package vessel temperature of 360.4°F under an HAC fire.

Auto-ignition is still very unlikely or not significant for papers with an auto-ignition point of 424~474°F, i.e., below the local payload temperature of 552°F, due to their negligible weight when compared with the allowed payloads. Auto-ignition, from papers with a weight load less than 0.1%, is not a significant safety concern for the package during transportation.

Radioactive contents have melting points above 600°F and there is no melting of the radioactive contents at the HAC maximum temperature of payload of 552°F

Temperatures inside the containment during an HAC fire may exceed the melting point of the hydrogenous packaging material. The potential melting of the packaging hydrogenous material will be limited and its impact negligible.

No interactions among the various contents and between the contents and the packaging materials.

Radioactive payload is a stable solid with a melting point above 600°F

Pressure increase due to the melting of packaging materials during an HAC fire can be vented to the atmosphere to avoid over-pressurization.


Gases generated in an HAC fire can be vented and released to the atmosphere through the plastic vent plugs which will melt in a fire event prior to yielding of the steel containment vessel.

No significant affect on the containment system of this Type AF package.

Note that the BU-D has plastic vent plugs that do not vent the containment system. There is no venting of the inner containment to atmosphere. The statement in the NRC SER refers to pressure generated by thermal degradation of insulating material between the inner containment and outer drum. A 1/8" thick high temperature heat resistant silicone coated fiberglass gasket is used between the steel flange ring and blind flange. The payload vessel blind flange is secured with twelve bolts. There are no penetrations, valves or venting devices used within the containment boundary [2].

Current Need

There is a need to allow greater than 200 grams of plastic material to allow packaging the contents in polyethylene containers that would be loaded in the Versa-Pac inner containment. After discussing a proposal to modify the 200-gram limit on plastic, the NRC staff provided the following comments [4]:

| | | |
|---|-----------------------------|---------------------|
|  | NOT FOR PUBLIC DISTRIBUTION | CN-13002-301 Rev. 1 |
| | | Page 7 of 26 |

- (I) *it might be wise to still keep a bounding value for plastics, possibly based on the criticality condition for hydrogen density greater than 0.141g/cm³ because contents are not well defined;*
- (ii) *contents could potentially be better defined with air void spaces between bottles being filled;*
- (iii) *the applicant should look at the effect of pressure buildup on the structural analysis of the package;*
- (iv) *the impact of toxic and corrosive gases on cavity materials and the seals must be evaluated even if staff recognizes that containment is not an issue in a Type AF package;*
- (v) *the applicant will need to demonstrate by calculation why auto-ignition is not a credible event (the current statement on the thermal protection of the package is insufficient), and why there is no safety concern with an unlimited amount of plastics; and*
- (vi) *the calculations should consider a range (small to large) of plastic and paper quantities with the available oxygen.*

Staff said that generic statements on a “negligible pressure increase” or on the “unlikelihood of auto-ignition” will not be accepted and that a rigorous justification of all assumptions is required for this amendment request to have any assurance of success.

The package is required to withstand a 30-minute fire without rupture of the inner container in order to contain the fissile contents. The external fire is the primary thermal event that may result in a secondary thermal event by auto-ignition of the contents or packing materials. Auto-ignition occurs at higher temperatures than flame ignition, and may occur after the external fire is extinguished due to the heat capacity of the contents and packaging materials.

2. METHOD OF ANALYSIS

2.1 Thermal degradation

As defined in NFPA 921, the ignition energy of a material is the quantity of heat energy per unit exposed surface area that must be absorbed by the material in order to pyrolyze, ignite, and burn. This energy is the product of the heat flux absorbed by the material and the time of exposure until ignition. A material with a given ignition energy will ignite faster if exposed to a high incident heat flux and slower if the incident heat flux is low. The amount of energy required for ignition also depends on the physical and chemical properties of the material, especially its thermal inertia and ignition temperature. However, the heat flux exposure must be greater than a certain critical value. For a heat flux less than or equal to this value, the ignition time is effectively infinite; i.e., ignition will not occur. This is the definition of the critical heat flux for ignition [5].

Combustion of contents or packing materials may result in formation of gases and temperature increase that could degrade the integrity of the inner containment. Temperature increase is associated with the energy from exothermic combustion reaction that consumes oxygen and fuel in the form of contents and packing materials. There may be increase or decrease in the gas molecules dependent on the stoichiometry of the combustion. In some cases, combustion reaction will decrease the total number of gas molecules. Cooling of the container will condense water vapor and result in a decrease in the pressure once the container returns to room temperature. The pressure increase is a transient condition that reaches a maximum and may decrease to a value less than the initial pressure before the combustion.

The inner container is sealed during loading at room temperature and pressure. The quantity of oxygen available to support combustion is limited by the normal pressure and volume inside the inner container. The volume is reduced by any contents and packing material with a maximum possible being the volume of the empty inner container.

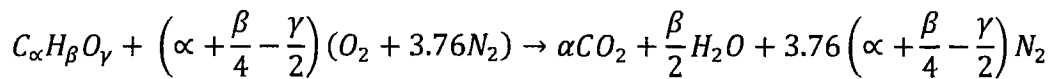
Once the ignition temperature is reached, the exothermic chemical reaction of combustion generates heat energy that is transferred to the contents and packaging materials. Heat generated by combustion is limited by either the amount of fuel material or oxygen available to sustain the chemical reactions. Once combustion starts in a sealed container the thermal degradation will progress to a point that the heavier CO₂ gases displace the O₂ and smother the flame.

2.2 Thermodynamics of combustion

The thermodynamics of combustion must be considered in order to evaluate the heat generation and subsequent pressure increase in a sealed container. Thermodynamics of combustion is quantified by the stoichiometry and heating value of the exothermic chemical reactions [6].

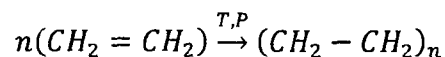
Stoichiometry of Combustion

A stoichiometric mixture contains the exact amount of fuel and oxidizer such that after combustion is completed, all the fuel and oxidizer are consumed to form products. This ideal mixture approximately yields the maximum flame temperature, as all the energy released from combustion is used to heat the products. Combustion stoichiometry for a general hydrocarbon fuel, $C_\alpha H_\beta O_\gamma$, with air can be expressed as



The amount of air required for combusting a stoichiometric mixture is called stoichiometric or theoretical air. The above formula is for a single-component fuel and cannot be applied to a fuel consisting of multiple components. A method of balancing systems with multiple fuels is to first develop stoichiometry relations for each fuel component individually, then, multiply the individual stoichiometry equations by the mole fractions of the fuel components and add stoichiometric combustion equations together.

Polyethylenes are thermoplastic materials that are produced primarily by the catalytic polymerization of ethylene gas (C_2H_4) at elevated temperatures (T) and pressures (P),

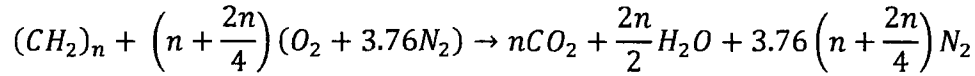


The polyethylene molecules usually contain branches of various lengths. Of all the hydrocarbon polymers, polyethylene has the simplest structure and the highest ratio of hydrogen to carbon in its backbone. Most commercial polyethylene formulations also contain additives, such as stabilizers, antioxidants, crosslinking agents, slip and anti block agents, and fire retardants.

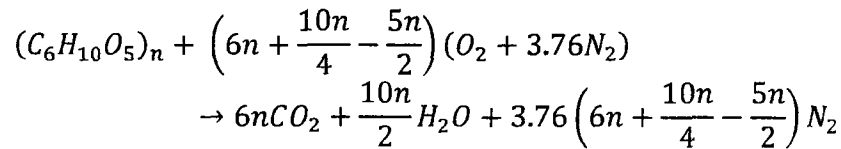
Thermoplastic materials, such as polyethylene and polypropylene, are commonly used as packing material for contents. Paper may also be present as labels on the contents containers. Paper is a mixture but its main constituent is cellulose, which is a complex carbohydrate with chemical formula $(C_6H_{10}O_5)_n$.

For example, the combustion stoichiometry for a fuel mixture that consists of paper or polyethylene materials is:

Polyethylene



Paper



Fuel and Air Content of Combustion Mixture

In practice, fuels are often combusted with an amount of air different from the stoichiometric ratio. If less air than the stoichiometric amount is used, the mixture is described as fuel rich. If excess air is used, the mixture is described as fuel lean. For this reason, it is convenient to quantify the combustible mixture using the following method:

Fuel-Air Ratio (FAR): The fuel-air ratio, f , is given by

$$f = \frac{m_f}{m_a}$$

where m_f and m_a are the respective masses of the fuel and the air. For a stoichiometric mixture, FAR becomes,

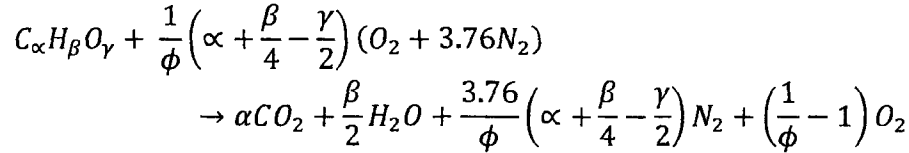
$$f_s = \frac{m_f}{m_a} = \frac{M_f}{\left(\alpha + \frac{\beta}{4} - \frac{\gamma}{2}\right) \cdot 4.76 \cdot M_{air}}$$

Equivalence Ratio: Normalizing the actual fuel-air ratio by the stoichiometric fuel-air ratio gives the equivalence ratio, ϕ .

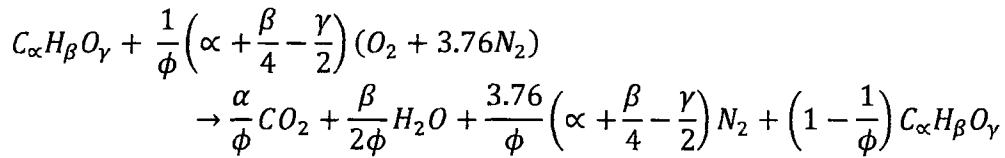
$$\phi = \frac{f}{f_s} = \frac{m_{as}}{m_a} = \frac{N_{as}}{N_a} = \frac{N_{O_2,s}}{N_{O_2,a}}$$

The subscript, *s*, indicates a value at the stoichiometric condition

Assuming that the products contain major species only (complete combustion) and excess air, the global equation for lean combustion $\phi \leq 1$ is



For rich combustion ($\phi > 1$), the products may contain CO, unburned fuels, and other species formed by the degradation of the fuel. Often, additional information on the products is needed for complete balance of the chemical reaction. If the products are assumed to contain only unburned fuel and major combustion products, the corresponding global equation can be written as



The inner container of the package can be modeled as a sealed combustion chamber. The fuel, polyethylene or paper, is burned where the oxygen available for combustion is limited to the air sealed inside the container. The mass of the air sealed inside the container can be determined using the density of air and the volume of the container.

$$m_a = \rho_a \cdot V$$

The mass of fuel consumed depends on the mass of air available to support the stoichiometric combustion.

$$m_f = f_s \cdot m_a$$

The molecular mass of the polyethylene, $(CH_2)_n$, is approximated as

$$M_f = M_{(CH_2)_n} = n \cdot 14.0247.$$

The stoichiometric fuel air ratio for polyethylene is

$$f_{polyethylene} = \frac{m_{f,(CH_2)_n}}{m_a} = \frac{n \cdot 14.0247}{\left(n + \frac{2n}{4}\right) \cdot 4.76 \cdot M_{air}} = \frac{14.0247}{1.5 \cdot 4.76 \cdot 28.96} = 0.0678.$$

The molecular mass of the paper, $(C_6H_{10}O_5)_n$, is approximated as

$$M_f = M_{(C_6H_{10}O_5)_n} = n \cdot 162.1312.$$

The stoichiometric fuel air ratio for paper is

$$f_{paper} = \frac{m_{f,(C_6H_{10}O_5)_n}}{m_a} = \frac{n \cdot 162.1312}{\left(6n + \frac{10n}{4} + \frac{5n}{2}\right) \cdot 4.76 \cdot M_{air}} = \frac{162.1312}{6 \cdot 4.76 \cdot 28.96} = 0.1960.$$

Heating Values

Heating values of a fuel (units of kJ/kg or MJ/kg) are traditionally used to quantify the maximum amount of heat that can be generated by combustion with air at standard conditions (STP defined as 25°C and 101.3 kPa). The amount of heat release from combustion of the fuel will depend on the phase of water in the products. If water is in the gas phase in the products, the value of total heat release is denoted as the lower heating value (LHV). When the water vapor is condensed to liquid, additional energy (equal to the latent heat of vaporization) can be extracted and the total energy release is called the higher heating value (HHV). The value of the LHV can be calculated from the HHV by subtracting the amount of energy released during the phase change of water from vapor to liquid as

$$LHV = HHV - \frac{N_{H_2O,p} M_{H_2O} h_{fg}}{N_{fuel} M_{fuel}} \text{ (MJ/kg)},$$

where $N_{H_2O,p}$ is the number of moles of water in the products. Latent heat for water at STP is h_{fg} (2.44 MJ/kg or 43.92 MJ/kmol). In combustion literature, the LHV is

normally called the enthalpy or heat of combustion (Q_c) and is a positive quantity.

The heats of combustion for commercial polymers of known chemical structure have been determined using an oxygen bomb calorimeter according to standard methods. The reported value for the heat of combustion for polyethylene is 47.7 kJ/g [7]. The heat of combustion for paper is 17.4 to 18.0 kJ/g [8].

The inner container can be represented by an adiabatic system where the starting temperature for the secondary combustion is the maximum temperature experienced during the external fire transient. The combustion products and steel material of the inner container absorb the heat energy from the combustion of plastic materials.

The amount of heat energy transfer to the combustion products and packaging materials is estimated by

$$Q = [\sum_i m_i c_{pi}] \Delta T .$$

The final temperature inside the container is estimated by

$$T_f = T_0 + \frac{Q}{\sum_i m_i c_{pi}} .$$

The heat energy transfer, Q , is the sum of the product of heat of combustion, Q_c , for the fuel materials and the mass of fuels, m_f , consumed by the combustion reaction.

$$Q = \sum_i Q_{ci} \cdot m_{fi}$$

The temperature in the sealed inner container will be greatest when there are no heat losses to the package materials (steel container and contents) and all of the energy released from combustion is used to heat the product gases. An increase temperature of gaseous combustion products inside the inner container will cause an increase in the pressure. The gaseous combustion products are assumed to behave as an ideal gas, where

$$PV = nRT .$$

The volume of the inner container is constant; therefore, any change in the number of moles of gases or change in temperature will have an effect on final pressure. Condensation of water vapor, a product of combustion, during the cool down phase of the thermal transient will decrease pressure inside the sealed inner container.

$$P_f = P_0 \frac{n_f T_f}{n_0 T_0}$$

2.3 Chemistry of Combustion

Cellulosic materials, such as wood or paper, decompose directly to gaseous vapors when heated, leaving behind a residue. Thermoplastics such as polypropylene or polyethylene undergo a two-step pyrolyzation process. As the thermoplastic is heated, it melts and turns into a liquid, and then this liquid melt is vaporized into the gaseous fuel. In an inert atmosphere, polyethylene begins to decompose at 290°C (565°K). Piloted ignition of polyethylene has been observed at a surface temperature of 368°C (640°K). The products of decomposition include a wide range of alkanes and alkenes. Decomposition is enhanced in air, with significant effects detectable at 151°C (423°K) [9]. Auto-ignition of polyethylene occurs generally at 330°C – 410°C depending on an individual product composition. The auto-ignition temperature of paper is 450°C (842°F).

When polyethylenes are thermally degraded under oxidative experimental or real fire conditions complex volatile organic chemical species are produced. The reaction to form these organic volatiles occurs partially within the heated polyethylene material, which has a restricted oxygen supply, and partially in the gas phase, which has a rich oxygen supply. Short-chain compounds, such as aldehydes and ketones, are generated predominantly by the gas-phase oxidation of the volatilized hydrocarbons.

When polyethylenes are exposed to elevated temperatures under inert atmospheric conditions, saturated and unsaturated hydrocarbons of various chain lengths are generated. Since polyethylenes do not contain any oxygen in their molecular structure, no CO, CO₂ or oxygen containing organic compounds are produced. The composition of the volatile compounds, which are formed from polyethylene products, will reflect the presence of any additives present in the formulation. Below 800°C the thermal degradation proceeds by a fragmentation process producing alkane and alkene type compound each containing 2 to 7 carbon atoms. At higher temperatures, degradation proceeds by formation of free radicals that unzip producing the ethylene monomer. The thermal degradation products of commercially available polyethylene in inert atmosphere are methane, ethylene, acetylene and benzene.

The following Degradation products are generated from polyethylene during pyrolysis (thermal degradation in inert atmospheres), thermooxidation (thermal degradation in oxidative atmosphere) and combustion (flaming degradation) [10]:

| Product | Pyrolysis (%) | Thermo-oxidation (%) | Combustion (%) |
|--|------------------|-------------------------|-------------------|
| Carbon dioxide | 0.1 | 0.4 | N |
| Propylene | 0.1 | 0.35 | 0.5 |
| Butylene | 4.5 | 3.7 | 2.7 |
| Butane | 1.5 | - | 2.3 |
| 1,3 Butadiene | 0.75 | 4.1 | 1.1 |
| Pentene+Propanal | 6.5 | 7.5 | 6.7 |
| - | - | 7.4 | - |
| n- Pentane | - | 6.2 | 3.9 |
| Butyraldehyde | - | 14.5 | 5.8 |
| Cyclohexane | - | 4.5 | 3.3 |
| Hexene-1 | 9.9 | - | 10.8 |
| n-Hexane | 4.8 | - | 4.9 |
| Cyclohexene | - | 0.27 | 2.2 |
| Benzene | 2.6 | 0.65 | 5.9 |
| Methylcyclopentene | - | 2.5 | 0.8 |
| Valeraldehyde | - | 11.5 | 5.6 |
| Heptene-1 | 5.6 | 1.4 | 3.0 |
| n- Heptane | 6.2 | 1.3 | 1.6 |
| Toluene | 0.8 | 0.9 | 3.3 |
| Ethylcyclopentene | 0.5 | - | - |
| Methyl isobutyl ketone (or allyl acetate) | - | 2.2 | 1.7 |
| Hexanal | - | 5.6 | 2.7 |
| Octen-1 | 5.7 | - | 3.9 |
| n-Octane | 4.6 | - | 2.0 |
| Acrylic acid | - | 0.9 | 0.8 |
| Ethyl benzene | 1.3 | 0.89 | 2.5 |
| Heptanal | - | 3.1 | 3.1 |
| Nonene-1 | 5.4 | 0.7 | 2.7 |
| n-Nonane | 3.5 | 1.4 | 1.2 |
| Epoxide | - | 0.3 | 0.2 |
| Keto-aldehyde | - | 0.6 | 0.3 |
| Octanal | - | 2.5 | 0.2 |
| Decene-1 | 5.8 | - | 0.7 |
| n-Decane | 3.0 | 0.6 | 0.3 |
| Undecene-1 | 4.3 | - | 0.9 |
| n-Undecane | 3.0 | - | 0.3 |
| Nonanal | - | 1.9 | - |
| Decanal | - | 0.8 | - |
| Tridecene-1 | - | 1.9 | 0.2 |
| n-Tridecane | 1.5 | - | 0.4 |
| Undecanal | - | 0.4 | - |
| Silicone | - | 0.1 | - |
| Tetradecene-1 | 2.0 | - | 0.15 |
| n-Tetradecane | 1.2 | - | 0.2 |
| Dodecanal | - | 0.2 | - |
| Pentadecene-1 | 1.8 | - | 0.15 |
| n-Pentadecane | 0.7 | - | 0.15 |
| Tridecanal | - | 0.1 | - |
| Hexadecene- 1 | 1.3 | - | 0.07 |
| n- Hexadecane | 0.9 | - | 0.07 |
| Tetradecanal | - | 0.1 | - |
| Heptadecene-1 | 0.5 | - | 0.15 |
| + n-Heptadecane | 0.4 | - | - |
| Pentadecanal | - | 0.1 | - |
| Octadecene-1 | 0.6 | - | 0.15 |
| + n-Octadecane | 0.5 | - | - |

3. REFERENCES

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- 2 Versa-Pac Safety Analysis Report, Century Industries, Bristol, VA.
- 3 NRC SAFETY EVALUATION REPORT, Model No. Versa-Pac Package, Certificate of Compliance No. 9342, Revision No. 6, February 2, 2012.
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4. DESIGN INPUT

Versa-Pac Safety Analysis Report [2]

Material data sheet for gasket [11]

5. ACCEPTANCE CRITERIA AND DESIGN ASSUMPTIONS

5.1 Acceptance Criteria

The integrity of the inner containment is maintained during accident fire conditions such that no fissile contents escape. This performance criteria is ensured by the following criteria:

Pressure in the inner container remains below the design pressure of 15 psig (29.7 psia) at STP[2].

The products of combustion do not degrade the inner containment lid seal such that air could be drawn into the container.

5.2 Design Assumptions

Heat flux exposure is greater than the critical heat flux for ignition for polyethylene and paper, even though the Versa-Pac thermal evaluation predicts surface temperatures on the inner containment that are less than the auto-ignition temperature polyethylene or paper.

Oxygen available for combustion is limited to that contained in the empty volume of the inner container at 25°C (298°K) and standard atmospheric pressure (P_{298K} = 14.7 psia). The actual volume of air will be less depending on the volume occupied by the contents and packing materials.

Complete stoichiometric combustion occurs until all the oxygen is consumed.

Pressure increase due to change in temperature or the number of gas molecules follows the ideal gas law.

Water produced by combustion vaporizes, therefore, the lower heating value (LLH) may be used to estimate temperature increase.

The maximum temperature of the inner container is assumed to be the maximum calculated weighted payload temperature of 360.4°F (182.4°C or 455K) at 22 minutes into the HAC fire. Although this is well below the auto-ignition temperatures of 424~474°F for paper, 660-986°F for plastics, it is assumed that combustion of any paper or plastic in the container occurs for the purpose of estimating the effect on pressure.

6. CALCULATION

The steel inner container of the Versa-Pac has a mass of 45 kg and the volume is approximately 0.08 m³. The container is sealed at room temperature and pressure (25°C, 101.3 kPa). The design pressure for the inner containment is 15 psig. The contents of the container is a solid material in polyethylene bottles with paper labels. The volume of air available for combustion is assumed to be equal to the volume of the steel container, although the actual volume would be less since the contents would displace air in the container. The method described Section 5.0 can be used to determine the pressure inside the Versa-Pac steel container prior to combustion of polyethylene and paper, the maximum pressure expected during combustion, and the final pressure after cooling down to room temperature.

6.1 Mass of fuel consumed with a limited air supply

The mass of air available for combustion of fuel materials is

$$m_a = \rho_a \cdot V = 1.25 \frac{\text{kg}}{\text{m}^3} \cdot 0.08 \text{ m}^3 = 0.1 \text{ kg}$$

The stoichiometric fuel-to-air ratio is used to calculate the mass that can undergo combustion as

$$m_{\text{polyethylene}} = f_{\text{polyethylene}} \cdot m_a = 0.0678 \cdot 0.1 \text{ kg} = 0.00678 \text{ kg}$$

$$m_{\text{paper}} = f_{\text{paper}} \cdot m_a = 0.1960 \cdot 0.1 \text{ kg} = 0.01960 \text{ kg}$$

6.2 Heat energy generated by combustion

The heat energy, Q , may be attributed either combustion of the polyethylene, paper, or some combination of both. In this example, there is not enough oxygen for combustion of all the paper or polyethylene. Each fuel type will be considered individually to bound the range of heat energy generated.

$$Q_{\text{polyethylene}} = Q_{c,\text{polyethylene}} \cdot m_{f,\text{polyethylene}} = 47.7 \frac{\text{kJ}}{\text{kg}} \cdot 0.00678 \text{ kg} = 0.3234 \text{ kJ}$$

$$Q_{\text{paper}} = Q_{c,\text{paper}} \cdot m_{f,\text{paper}} = 18 \frac{\text{kJ}}{\text{kg}} \cdot 0.01960 \text{ kg} = 0.3528 \text{ kJ}$$

6.3 Thermal mass of packing material or combustion products

The thermal mass is the mass of a material, m , times the specific heat capacity, c_p . For a mixture of numerous different materials, the thermal masses for the different components can just be added together. The thermal mass is used to calculate the temperature change caused by a heat energy generated by combustion.

The specific heat of steel c_p (J/kg K) can be determined by [12]:

$$c_p = \begin{cases} 425 + 7.73 \times 10^{-1}T - 1.69 \times 10^{-3}T^2 + 2.22 \times 10^{-6}T^3 & \text{for } 20^\circ\text{C} \leq T < 600^\circ\text{C} \\ 666 + 13002/(738 - T) & \text{for } 600^\circ\text{C} \leq T < 735^\circ\text{C} \\ 545 + 17820/(T - 731) & \text{for } 735^\circ\text{C} \leq T < 900^\circ\text{C} \\ 650 & \text{for } 900^\circ\text{C} \leq T < 1200^\circ\text{C} \end{cases}$$

The specific heat capacity of steel at 330 C is 0.576 kJ/kg-K, the specific heat of the nitrogen and carbon dioxide is 1.075 kJ/kg K, and the specific heat of water vapor is 2.105 kJ/kg K. The mass of each combustion products (N_2 , CO_2 , and H_2O) can be estimated from the stoichiometric ratio to air as

$$\frac{m_p}{m_a} = \frac{N_p \cdot M_p}{\left(\alpha + \frac{\beta}{4} - \frac{\gamma}{2}\right) \cdot 4.76 \cdot M_{air}}$$

where N_p and M_p

$$N_{CO_2} = \alpha, \quad M_{CO_2} = 44.01 \text{ kg/kmol}$$

$$N_{H_2O} = \frac{\beta}{2}, \quad M_{H_2O} = 18.02 \text{ kg/kmol}$$

$$N_{N_2} = \left(\alpha + \frac{\beta}{4} - \frac{\gamma}{2}\right) \cdot 3.76, \quad M_{N_2} = 28.02 \text{ kg/kmol}$$

and molecular mass of air is 28.97 kg/kmol.

For the air mass of 0.1 kg, the mass of the combustion products for polyethylene or paper is estimated as,

| | Paper, (C ₆ H ₁₀ O ₅) _n | Polyethylene, (CH ₂) _n |
|--|--|---|
| N _{CO2} (moles) | 6 | 1 |
| m _{CO2} (kg) | 0.03192 | 0.02128 |
| N _{H2O} (moles) | 5 | 1 |
| m _{H2O} (kg) | 0.01089 | 0.00871 |
| N _{N2} (moles) | 22.56 | 5.64 |
| m _{N2} (kg) | 0.07640 | 0.07640 |
| Σ _i m _i c _{pi} (kJ/K) | 0.1394 | 0.1233 |

6.4 Pressure rise due heat transfer from outside the containment

The final pressure due to the rise from room temperature, $T_o=298$ K, to the maximum calculated wall temperature of the inner container, $T_{f1}=455$ K, is estimated as

$$P_{455K} = P_{298K} \frac{T_{f1}}{T_o} = 14.7 \text{ psia} \frac{455 \text{ K}}{298 \text{ K}} = 22.4 \text{ psia}$$

The temperature, T_{f2} , and pressure, P_{f_poly} , after complete combustion of polyethylene, assuming an adiabatic system where all the heat is absorbed by the gas products inside the inner container is estimated as

$$T_{f2} = T_{f1} + \frac{Q_{polyethylene}}{\sum_i m_i c_{pi}} = 455 \text{ K} + \frac{0.3234 \text{ kJ}}{0.1233 \text{ kJ/K}} = 458 \text{ K}$$

$$P_{f_poly} = P_{o,455K} \frac{n_{f_poly} T_{f2}}{n_{o_poly} T_{f1}} = 22.4 \text{ psia} \frac{7.64 \text{ mol } 458 \text{ K}}{7.14 \text{ mol } 455 \text{ K}} = 24.1 \text{ psia}$$

Likewise, assuming only paper combustion occurs, the final pressure, due to the rise from room temperature to the maximum calculated wall temperature of the inner container, $T_{f3}=455$ K, is estimated as

$$P_{455K} = P_{298K} \frac{T_{f3}}{T_o} = 14.7 \text{ psia} \frac{455 \text{ K}}{298 \text{ K}} = 22.4 \text{ psia}$$

The temperature, T_{f4} , and pressure, P_{f_paper} , after complete combustion of paper, assuming an adiabatic system where all the heat is absorbed by the gas products inside the inner container is estimated as

$$T_{f4} = T_{f3} + \frac{Q_{paper}}{\sum_i m_i c_{pi}} = 455 \text{ K} + \frac{0.3528 \text{ kJ}}{0.1394 \text{ kJ/K}} = 458 \text{ K}$$

$$P_{f_paper} = P_{455K} \frac{n_{f_paper} T_{f4}}{n_{o_paper} T_{f3}} = 22.4 \text{ psia} \frac{33.56 \text{ mol } 458 \text{ K}}{28.56 \text{ mol } 455 \text{ K}} = 26.5 \text{ psia}$$

The increase in pressure is due primarily to rise in temperature from room temperature to the auto-ignition temperature of the polyethylene or paper. The heat energy added by the combustion reaction adds only about 3K to the auto-ignition temperature due to the limited air supply inside the sealed container. The actual system would not be adiabatic, and the steel inner container would absorb heat energy from the combustion. The thermal capacity of steel in the temperature range of the combustion is 0.346 to 0.635 kJ/kg K. The temperature rise would be negligible due to the large thermal mass, if the 45 kg inner container absorbed the heat energy. In either case, the number of moles of gases increases due to combustion products and water in vapor form. The heat of combustion used is a lower heating value that accounts for the enthalpy of vaporization for water.

After cooling to the initial room temperature, the number of moles of gaseous product is reduced by the number of moles of water produced by the combustion, and a small amount of heat is released by the condensation of water. The final pressure, ignoring the heat of vaporization released, after cool down is estimated as

$$P_{f_paper_cooldown} = P_{298K} \frac{(n_{f_paper} - N_{H2O}) T_{f_cooldown}}{n_{o_paper} T_0} = 14.7 \text{ psia} \frac{(33.56 - 5) \text{ mol } 298 \text{ K}}{28.56 \text{ mol } 298 \text{ K}}$$

$$= 14.7 \text{ psia}$$

$$P_{f_poly_cooldown} = P_{298K} \frac{(n_{f_poly} - N_{H2O}) T_{f_cooldown}}{n_{o_poly} T_0} = 14.7 \text{ psia} \frac{(7.64 - 1) \text{ mol } 298 \text{ K}}{7.14 \text{ mol } 298 \text{ K}}$$

$$= 13.8 \text{ psia}$$

The Versa-Pac 110 gallon package has an inner container with nearly twice the mass and volume as the Versa-Pac 55 gallon package. The increase in volume would result in doubling the amount of air available for combustion, the paper or polyethylene that undergoes complete combustion, and the mole of product gases produced. The heat generation due to combustion inside the containment would double, but there would be twice the heat capacity ($\sum_i m_i c_{pi}$) in the product gases. Hence, the temperature and pressure increase due to combustion reaction is independent of the inner container volume.

6.5 Combustion required to exceed the design pressure

The amount of paper or polyethylene required to undergo complete combustion in order to exceed the design pressure can also be calculated. This assumes that there is the stoichiometric quantity of air available in the inner container to support the combustion. Also, the thermal mass of product gases is assumed to remain constant as calculated for 0.1 kg of air. This assumption underestimates the mass of air, because the thermal mass of combustion products is needed to calculate the mass of air. The thermal mass of combustion products is a function of the mass of air used for combustion of the fuel. An iterative calculation could be used to converge on the exact quantity of air, but the result would be a larger quantity of air than calculated by assuming constant thermal mass of product gases. The final pressure would be the design pressure of the inner container, 29.7 psia (15 psig + 14.7 psia). Starting with this pressure, and rearranging the formulas used previously used to calculate a final pressure, the mass of paper or polyethylene can be calculated. The final temperature is calculated using the ideal gas law, the heat energy input (Q) is calculated for the required to increase from the auto-ignition temperature (T_o) to the final temperature (T_f), and the resulting Q is used to calculate a mass of fuel material for the heat of combustion (Q_c).

$$T_{f_paper_combustion} = T_{f3} \frac{P_{design} n_{o_paper}}{P_{455K} n_{f_paper}} = 455 K \frac{29.7 \text{ psia } 28.56 \text{ mol}}{22.4 \text{ psia } 33.56 \text{ mol}} = 512 K$$

$$Q_{paper_combustion} = (T_{f_paper_combustion} - T_{f3}) \sum_i m_i c_{pi} = (512 K - 455 K) 0.1394 \text{ kJ/K}$$

$$= 8 \text{ kJ}$$

$$m_{f,paper} = \frac{Q_{paper_combustion}}{Q_{c,paper}} = \frac{8 \text{ kJ}}{18 \frac{\text{kJ}}{\text{kg}}} = 0.44 \text{ kg}$$

Based on the stoichiometric ratio for paper combustion reaction, the air required for combustion of 0.44 kg paper is

$$m_a = \frac{m_{f,(C_6H_{10}O_5)_n}}{f_{s,(C_6H_{10}O_5)_n}} = \frac{0.44 \text{ kg}}{0.1960} = 2.3 \text{ kg}$$

Likewise for polyethylene, the mass of paper and air required for combustion to result in exceeding the design pressure may be calculated.

$$T_{f_poly_combustion} = T_{f1} \frac{P_{design}}{P_{455K}} \frac{n_{o_poly}}{n_{f_poly}} = 455 \text{ K} \frac{29.7 \text{ psia}}{22.4 \text{ psia}} \frac{7.14 \text{ mol}}{7.64 \text{ mol}} = 563 \text{ K}$$

$$Q_{poly_combustion} = (T_{poly_combustion} - T_{f3}) \sum_i m_i c_{pi} = (563 \text{ K} - 455 \text{ K}) 0.1233 \text{ kJ/K} \\ = 13.3 \text{ kJ}$$

$$m_{f,polyethylene} = \frac{Q_{poly_combustion}}{Q_{c,polyethylene}} = \frac{13.3 \text{ kJ}}{47.7 \frac{\text{kJ}}{\text{kg}}} = 0.28 \text{ kg}$$

Based on the stoichiometric ratio for polyethylene combustion reaction, the air required for combustion of 0.28 kg polyethylene is

$$m_a = \frac{m_{f,(CH_2)_n}}{f_{s,(CH_2)_n}} = \frac{0.28 \text{ kg}}{0.0678} = 4.1 \text{ kg}$$

7. Conclusions and Recommendations

Combustion in a sealed container is limited by the amount of air present to support the chemical reaction for the thermal degradation of the fuel. The maximum temperature in the sealed container will determine the maximum pressure. The sealed inner containment of the Versa-Pac contains only enough air for combustion of approximately 0.02 kg of paper and 0.007 kg of polyethylene. In the case of fuel rich combustion, the design pressure of the Versa-Pac could be exceeded only if sufficient air were present for combustion of approximately 0.44 kg of paper or 0.28 kg of polyethylene. Limits on the quantity of combustible materials, such as paper and polyethylene, should be based on the maximum temperature increase that results from the fuel rich combustion of paper or polyethylene. The maximum temperature is limited by the quantity of air present in a sealed container. Realistically, there would be no combustion of paper or polyethylene since the maximum calculated wall temperature of the inner vessel is much lower than the autoignition temperature of both paper and polyethylene. Therefore, there is no need to limit the quantity of paper or polyethylene to ensure the integrity of sealed Versa-Pac inner container.

The volume of the sealed container limits the air available for combustion. The additional air necessary to exceed the design pressure would require that the container not be sealed. This amount of air is approximately 20 to 40 times (4.1 kg for paper and 2.3 kg for polyethylene) the mass of air that is available in the sealed container (0.1 kg of air). The quantity of paper or plastic that can undergo combustion with the 0.1 kg of air in the sealed container (0.02 kg of paper or 0.007 kg of polyethylene) is much less than that required to reach the design pressure of the inner containment (0.44 kg paper or 0.28 kg polyethylene).

A flat gasket made from fiberglass-reinforced silicone sheet provides the seal for the inner container lid. This gasket material is suitable for operating temperatures from -100 to 500F. During normal shipping conditions, the silicone containment seal is compatible with the approved contents and temperature conditions. Thermal degradation of polyethylene, or other thermoplastic materials included with the contents, produces a range of alkane and alkene compounds. Silicone materials in general have acceptable compatibility with the products resulting from thermal degradation of the thermoplastic materials. However, nitrile materials are recommended for service applications where the gasket is in contact with alkanes, and fluorocarbon materials are recommended for service applications where the gasket is in contact with alkenes [13]. The limited air available to support combustion inside the containment and the short duration of the external thermal event, limit the extent of thermal degradation that could produce alkanes or alkene products. The heat energy transferred from the external thermal event or added by

a secondary internal combustion of the plastic packing material would not produce sufficient thermal degradation products to compromise the integrity of the silicone in the gasket material.

The limit on the quantity of hydrogenous packing material in the Versa-Pac approval certificate, USA/9342/AF-96, should be removed, and the contents specified as:

5.(b)(1) Type and Form of Material

Contents may be pre-packaged in polyethylene, polytetrafluoroethylene, aluminum, and carbon steel per Table No.1-4 of the application. Aluminum Trihydrate, Sodium Borate (Borax, fused), perlite, paper labels, plastic tape, plastic bags, plastic bottles and desiccant such as "Quik-Solid®" are also authorized as packing materials. Materials with a hydrogen density greater than 0.141 g/cm³ are not authorized.

Radioactive contents shall have an auto-ignition temperature and melting point greater than 600°F.

A limit on the quantity of hydrogenous packing material has no safety significance for criticality safety or containment of the radioactive material contents.