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Thermal Test Driven Pressure Build-up Inside Type-B Packages Containing Wet Radioactive Waste

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ABSTRACT

In recent years several German approval procedures for ductile cast iron transport containers containing wet intermediate level waste were conducted. BAM, as one of the German competent authorities, was involved in the complex design assessment work with this specific issue. Thermal analysis is one part of the authority assessment work done by BAM in Germany.

The radioactive contents of package designs which were not dried, only drained, consist of saturated ion exchange resin and a small amount of free water. Compared to the safety assessment of packages with dry content, attention must be paid to some more specific points. The most interesting point, however, is the pressure build-up inside the package due to vaporization. This could be caused by radiolysis of the liquid and must be taken into account for the storage period. The inner pressure of the package leads to mechanical loads to the package body, the lid and the lid bolts. Thus, the pressure is the driving force on the gasket system regarding the activity release and a possible loss of tightness.

The paper deals primarily with the pressure build-up inside the package caused by the transport regulatory thermal test (30 min at 800 °C) as part of the cumulative test scenario under accident conditions of transport. The pressure build-up is estimated by calculation in a very conservative way regarding conduction and heat radiation. Furthermore the paper discusses a conservative approach for the estimation of the resulting pressure depending on the percentage of water inside the cask. To get trustworthy results without an exact specification of the content, experimental fire tests should be conducted.

However, this paper shows the difficulties of assessing casks containing wet content. From the authority assessment point of view, drying of the content could be an effective way to avoid the above described pressure build-up and the associated difficulties for the safety assessment.

INTRODUCTION

In Germany the mechanical and thermal safety assessment of approved packages for the transport of RAM is carried out by BAM as the competent authority according to the IAEA regulations. BAM was involved in several approval procedures with ductile cast iron containers containing wet intermediate level waste. Commonly these contents consist of saturated ion exchange resin and a small amount of free water. During handling the contaminated ion exchange resins are flushed into the package using water and afterwards the ion exchange resin is just drained, but not dried.

Compared to the safety assessment of packages with dry content, attention must be paid to some more specific points. The physical and chemical compatibility of the content itself and of the content with materials of the package must be shown. From the mechanical resistance approach the package has to withstand the forces resulting from the freezing liquid. The most interesting point, however, is the pressure build-up inside the package due to vaporization. This could be caused by radiolysis of the liquid and must be taken into account for the storage period.

The paper primarily deals with the pressure build-up inside the package caused by the regulatory thermal test (30 min at 800 °C) as part of the cumulative test scenario under accident conditions of transport. The total pressure, which is the sum of the internal pressure of the package in addition to the pressure resulting from the heat of the thermal test, has to be seen as the driving force in a mechanical point of view. The mechanical resistance parameters of the package (body, lid, gasket), which are influenced by the heat and the previous mechanical tests, must be compared to the driving force.

The procedure of vaporization is shown and two approaches to determine the partial pressure of water steam are discussed: On the one hand the use of a CFD/FE model with its uncertainties and on the other hand an analytical approach for which steam tables were put into graphs with relevant filling degrees to show the pressure build-up inside the package.

SPECIFIC ASPECTS OF SAFETY ASSESSMENTS CAUSED BY WET CONTENTS

Compared to dry contents, the use of wet contents leads to the need for special attention to regulatory points of the regulation SSR-6 [1] Para 639 and 614.

Para 639 states "...shall take into account temperatures ranging from -40 °C to +70 °C for the components of the packaging." And also: "Attention shall be given to freezing temperatures for liquids...". According to this paragraph the freezing of liquids and the resulting freezing pressure as a mechanical load for the packages components (in particular the gasket) must be taken into account.

Para 614 mentions, that "The materials of the packaging and any components or structures shall be physically and chemically compatible with each other and with the radioactive contents." It should be noted, that wet/liquid does not necessarily mean that the fluid part is water. On the one hand it must be taken into account that different parts of the content could react chemically with each other up to exothermal reactions or yet generate even more aggressive substances probably due to pyrolysis. On the other hand this paragraph means that simply corrosion of the packages and the gasket system must be prevented. Using huge amounts of fluids in the content ("liquid radioactive material") Para 649 must be followed. "The design of a package intended for liquid radioactive material shall make provision for ullage to accommodate variations in the temperature of the contents, dynamic effects and filling dynamics." Regarding wet contents like ion exchange resin this paragraph is not crucial.

The most interesting point, however, is the pressure build-up inside the package due to vaporization. This could be caused by radiolysis of the liquid and must be taken into account for a possible storage period as well. The generation of hydrogen is not considered here, but the maximum allowable rate (self-ignition) of hydrogen could lead to a limitation criteria of maximum liquid amount too. The pressure resulting from the radiolysis could be calculated regarding the amount of energy inside the content, which is released during a possible storage period. This procedure can be seen as conservative, because only a part of the released energy will be absorbed by the content itself generating radiolysis gas.

However, the highest load to the package due to pressure will be generated by the pressure build-up caused by the thermal test under accident condition of transport.

THERMAL TEST

According to Para 728 [1] a fully engulfing, 30 min. duration fire period with an average temperature of at least 800 °C should be applied to the package. As initial condition the package, usually with its impact limiter, shall be in thermal equilibrium under conditions of an ambient temperature of 38 °C. The decay heat and also the solar insulation must be considered. The fire period should be followed by a cooling down phase with the same ambient conditions like the initial conditions. The cooling down phase has to last until the temperatures will be decreasing at every part of the package.

VAPORIZATION OF WATER

The vaporization of the fluid (in this case water) is the main process regarding the pressure build-up caused by the thermal test. Vaporization is the phase transition from a liquid phase to the gaseous phase. At normal pressure (100 kPa) liquid water will not increase its temperature above 100 °C but vaporize. This phase transition requires high amounts of energy. The heating of 1 liter of water from 38 °C to 100 °C (liquid) requires a total energy of 260 kJ. The vaporization of 1 liter of water at 100 °C requires 2088 kJ, which is more than 8 times the energy used to heat it up from 38 °C to 100 °C. These values do not consider the additional work to be done to pressurize the gas. With an increasing ambient pressure, this amount of work rises in this isochoric process.

During the phase transition from liquid to gas the volume of water increases about 1600 times.

At a given temperature water will vaporize, until the partial water pressure reaches the specific vapor pressure. Fig. 1 shows the vapor pressure as a function of temperature [2].

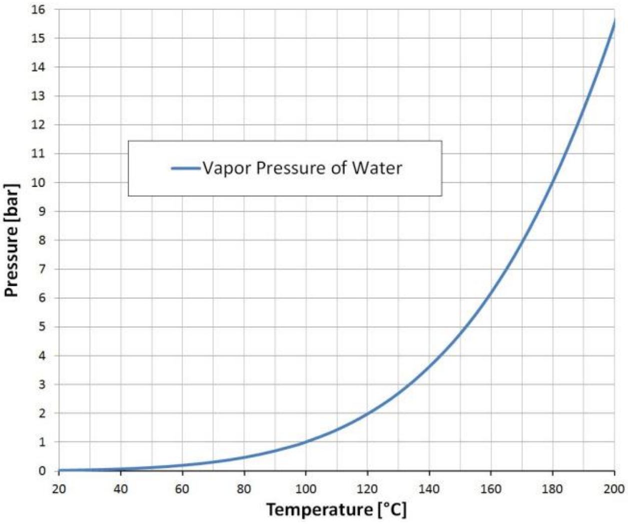

FIG. 1 VAPOR PRESSURE OF WATER AS A FUNCTION OF THE TEMPERATURE

Fig. 1 could also be read the other way: The boiling point of water depends on the ambient pressure. This means for example if the ambient pressure rises up to 2.7 bar (0.27 MPa) the water will be boiling at a temperature of 130 °C.

THE TOTAL PRESSURE INSIDE A PACKAGE

The package is stressed by the total pressure inside the package. On the one hand, this pressure leads to mechanical loads to the packages body, the lid and the lid bolting. On the other hand, the pressure is the driving force on the gasket system regarding the activity release and a possible loss of tightness.

The total pressure for any calculation is the sum of partial pressures of different gases like “air”, helium, water vapor or hydrogen.

The following effects could increase the total pressure:

a. Initial fill up of the packages with an inert gas like helium during handling at normal temperature P_0
b. Increasing pressure P_1 due to warming up to 38 °C as stated in [1]
c. Increasing pressure P_2 due to heating up caused by decay
d. Pressure build-up P_3 caused by radiolysis/chemical processes for the hole storage period
e. Vaporization of water P_4 driven by temperature at normal conditions of transport
f. Vaporization of water P_5 driven by temperature at accident conditions of transport
g. Pressure P_6 resulting from combustion and pyrolysis

Under normal conditions of transport the total pressure results to:

$$P_{tot,NCT} = P_0 + P_1 + P_2 + P_3 + P_4 \leq 700 \text{ kPa}$$

Under accident conditions of transport the total pressure results to:

$$P_{tot,ACT} = P_0 + P_1 + P_2 + P_3 + P_5 + P_6$$

LOCAL EFFECTS INSIDE PACKAGES DUE TO RAPID HEATING

Contrary to common models of the process of vaporization in a closed system (autoclave e.g.) an uneven heating of the wet content occurs in packages with thick walls. A thermal wave spreads out towards the center of the container and its content during the fire test and even in the cooling down phase (see fig. 3). As one consequence the maximum input of energy into the content happens after the fire period. At second the areas of the content close to the inner wall of the cask will be hotter than the inner areas. This means that in the areas the vapor pressure will be exceeded earlier and the free as well as the physically bound water starts to vaporize. Since the inner area of the content is cooler, the present steam condenses in this area. This results in a steam flow from the hotter areas into the cooler areas of the content. This steam flow transports mass in terms of water vapor stream, however also certain amounts of thermal energy (enthalpy). Thus a lowering of temperature happens in the outer sections (as in the case of evaporative cooling) and inner sections will be heated up due to condensation. In total this process tends to balance the temperatures of the contents. Since the steam produced moves towards cooler areas and condenses, a lower pressure than expected is being build up.

This process of balancing the temperatures and with this the reduction of the pressure build-up depends on many boundary conditions. The availability of free and physically bound water for vaporization depends on water permeability and bonding force of the content.

The progression of a steam flow is strongly depending on the temperature distribution and the resistance to the flow. The more or less generic description of the content by the applicant (for such kind of transport approval procedures) due to insufficient availability of information does not allow inference of the above mentioned boundary conditions.

DISCUSSION OF TWO APPROACHES FOR DETERMINATION OF THE PRESSURE BUILD-UP

For safety assessment (and even for the design/dimensioning of the package) it is necessary to determine the resulting total pressure inside the package under every possible condition regarding the transport regulations. In general the crucial condition regarding the pressure build-up due to vaporization is the thermal test as a part of the accident conditions of transport. For a package including adequate described content there are two ways to determine the pressure build-up besides the possibility of carrying out an experimental fire test:

The first option is to set up a CFD (Computational Fluid Dynamics) or a FE (Finite Element) model. A CFD model is capable of implying a fluid flow e.g. free convection. Considering radiation and heat conduction (even in a fluid) only and neglecting the fluid flow, a FE model would be sufficient.

The second option is the use of steam tables to determine the pressure build-up. This requires the knowledge of the amount of free water and free volume inside the package. Additionally a uniform temperature over the whole content (solid + water + gas) must be stated in a conservative way.

Both options will be described briefly and demonstrated by an example in the following.

CFD MODEL FOR THE DETERMINATION OF THE PRESSURE BUILD-UP

The following discussed modeling including the physical effects and boundary conditions refer to the generic cask shown in fig. 2. This cask contains ion exchange resin in combination with water as fluid.

The model cask was designed as a ductile cast iron cylinder with 2 m in height and an outer diameter of 1 m. The interior space is 1.5 m high and the inner diameter is 0.5 m. The interior space is filled with wet ion exchange resin up to a height of 1.10 m. Since the wet ion exchange resin is not specified properly its thermal properties are defined as a combination of the properties of water (60%) and air (40%). An air-filled cavity height of 0.40 m is considered above the content. The base and the lid-area of the cask are covered by encapsulated wooden impact limiter and the cask stands upright. As initial condition a temperature of 38 °C (acc. to IAEA SSR-6 [1]) is assumed for the complete cask since the decay heat in this kind of content is usually very low.

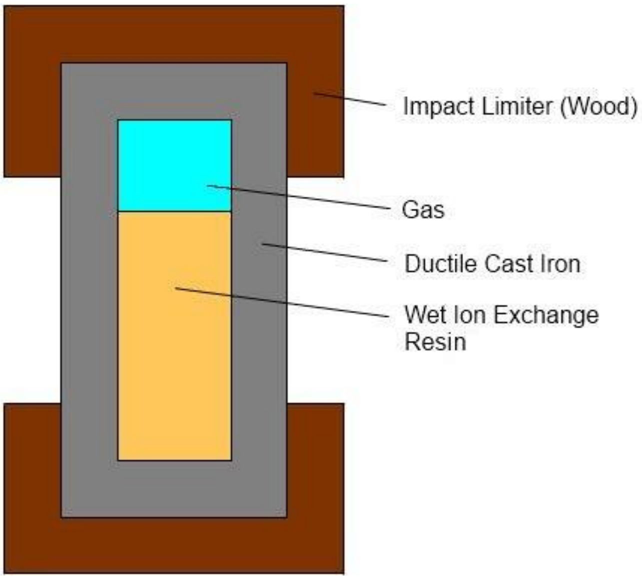

FIG. 2 GENERIC MODEL OF A CASK USED TO CALCULATE TEMPERATURES

The numerical model of the above described cask was performed in the fluid-dynamic code ANSYS® CFX® [3]. All materials of the cask and the wet ion exchange resin were modeled as solids, only the air-filled cavity was considered as a flow region including convection. The thermal properties of the ion exchange resin are assumed as the above described mixture of water and air. For all other materials their real conductivities, thermal capacities and densities were defined. The analysis was performed as a transient calculation with time steps of 15 seconds during the fire period and 30 seconds in the cooling down phase over a timeframe of 10 hours.

The consideration of the heat transport into the content is one possibility if a CFD/FE model is used which considers conduction and radiation only. The thermal energy evaluated in this way has to be related to the content.

With the above described modeling the temperature of the content right at the cask wall will be calculated noticeably too high. In reality the boiling water remains at the temperature of 100 °C until the local water is vaporized entirely. Due to the modeling the temperatures at the interface of cask and content exceeds 100 °C which leads to a lower thermal gradient. This results in an underestimation of the thermal energy absorbed. Additionally gasification or even pyrolysis of content is not considered with this simplified modeling.

The evolution of the temperature achieved from the numerical calculations is shown in fig. 3. The heat flux by time is analyzed in fig. 4 to consider the thermal energy which is transported into the content.

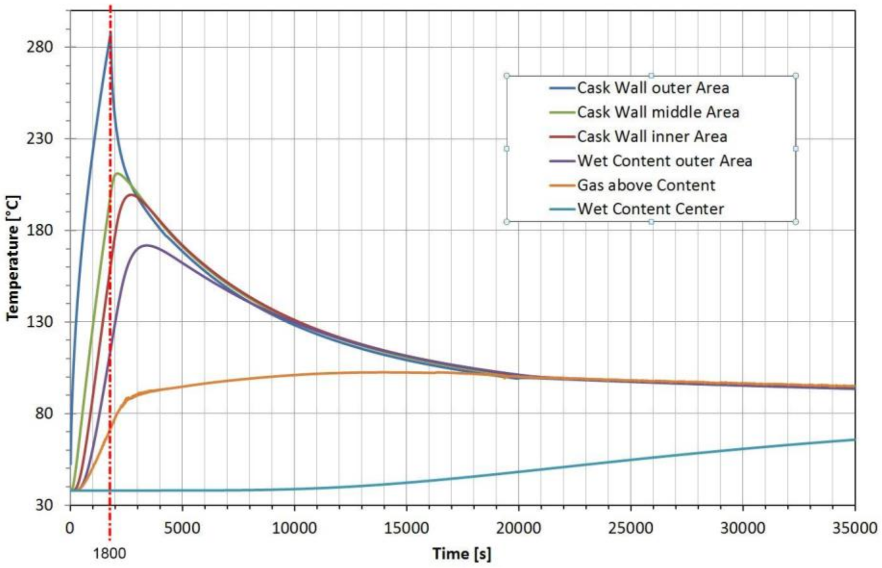

FIG. 3 TEMPERATURES IN THE CASK DURING THE 30 min (1800 sec) FIRE PERIOD AND THE FOLLOWING COOLING DOWN PHASE

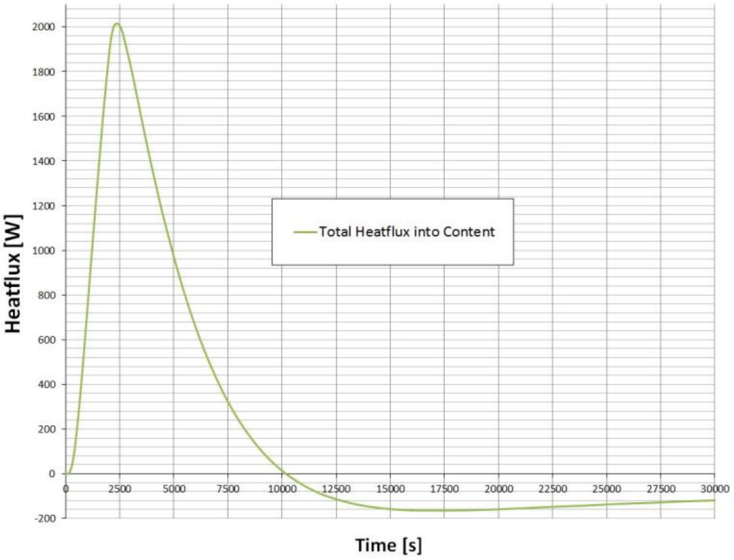

FIG. 4 HEAT FLUX INTO CONTENT AS A FUNCTION OF TIME

The entire transported thermal energy into the content can be achieved by integration of the heat flux over time and results to a maximum of 8127 kJ. The isothermals in the model cask at the point of the highest heat flux are shown in fig. 5.

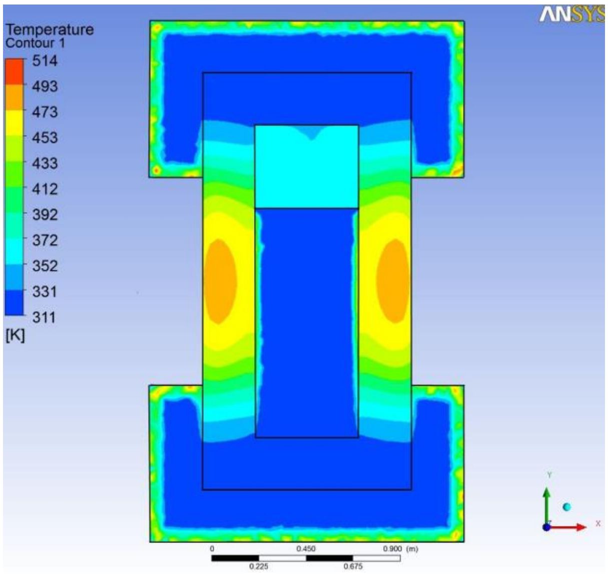

FIG. 5 CROSS SECTION OF THE CASK MODEL WITH ISOTHERMALS AT THE TIME OF THE MAXIMUM HEAT FLUX INTO THE CONTENT

Since it is not possible to establish a proposition about the pressure based on the temperatures, additional consideration is given to energy. Concerning the simplified and noticeably not conservative assumptions a thermal energy of 8127 kJ was transported into the content at the point t = 10000 s (based on the initial temperature of 38 °C). Due to the insufficient specification of the content and the uncertainties in the numerical calculations a heating of the content in a layer of 5 mm at the cask wall will be examined with respect to the evaluation of a cask. This layer means a volume of 0.0043 m^3 which contains 2.6 liters of water. With the available energy the assumed amount of water will be vaporized to 4100 liters of steam. Since the inner volume of the cask (pores in the resin and the entire cavity) is 0.208 m^3, the water vapor partial pressure will be approx. 2 MPa (20 bar) in a first assumption. Actually partial pressures resulting from the emergence of gas out of radiolysis and additional effects are not taken into account so far. Even though the considered energy seemed to be rather small, a positive assessment is not possible for this simplified model.

In the thermal assessment the description of the content often covers everything, which means that the maximum masses and maximum radioactive activity of the content are given. In the operation of casks also a partly filling of a container is possible, e.g. at the end of the loading campaign. Assumed that such a container is filled with 5 liters of ion exchange resin only, a similar calculation like discussed before can be done: The small amount of water vaporizes completely, in this case into the larger space of 0.294 m^3 for gas. The resulting pressure will be approx. 1.5 MPa (15 bar) solely from the water vapor partial pressure.

In summary it can be shown by reference to this model cask, that a pressure of far more than 2 MPa (20 bar) can be achieved actually with simple and not even conservative assumptions.

USE OF STEAM TABLES FOR THE DETERMINATION OF THE PRESSURE BUILD-UP

Vaporization is the transition from a liquid phase to the gaseous phase. This is shown in the water steam diagram in fig. 6 which is plotted over the temperature and the entropy. The diagram is constructed with values from [2].

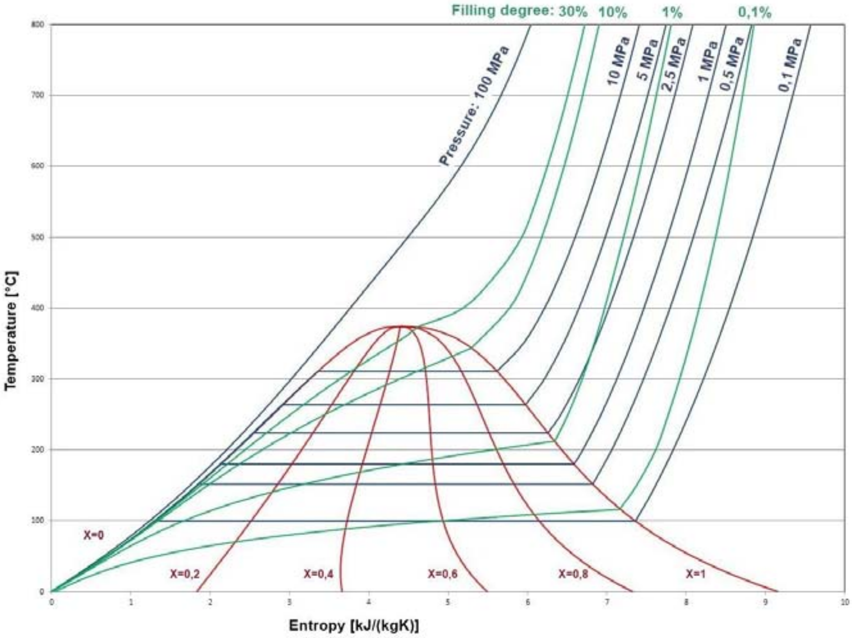

FIG. 6 TEMPERATURE ENTROPY DIAGRAM SHOWING THE DEPENDENCY OF THE FILLING DEGREE, THE POTENTIAL PRESSURE BUILD-UP AND THE TEMPERATURE

Used for the determination of the pressure build-up inside a package the diagram shows the correlation between the filling degree, a uniform temperature of the content and the resulting water steam pressure.

In the diagram the green lines are plotted for different filling degrees of the cask. The blue curves show isobars of the fluid (water, steam or superheated steam). The red boiling curve (X=0) is partly overlapped by isobars in the diagram, and meets the saturated liquid phase boundary (X=1) at a critical temperature of 374.12 °C. Between these curves the wet steam region is located.

In a closed cask, the heating up to a chosen temperature is an isochoric process. In fig. 6 this process is described in dependence of the filling degree F_D, which is the ratio between the volume of water V_W in the cask and the sum of the volume of water and the free volume V_F which is not taken by any other content [$F_D = V_W/(V_W+V_F)$]. With a given filling degree one has to follow the according green line from the point of origin up to the chosen temperature. The temperature and the partial pressure rise in the wet steam. Even if all water in a cask is evaporated (X=1), the partial pressure will still rise with a continuing application of energy.

As an example the state of the container shown in fig. 2 will be investigated. Assuming that the yellow marked ion exchange resin consists of 40% air, 30% solid and 30% water, a filling degree of $F_D = 28\%$ is easily reached. Additionally a conservative temperature for the whole content has to be chosen. Due to a lack of information concerning the content temperature, a temperature of the content has to be assumed. Therefore the maximum cask wall inner area temperature of approx. 200 °C from fig. 3 will be used. Using this temperature and a filling degree F_D of 30% the pressure taken from fig. 6 results to approx. 1.7 MPa (17 bar).

Evaluating the way to determine the pressure build-up using steam tables the following is easy to see: The need for choosing a conservative temperature for the content is the most crucial thing. One has to act on the assumption that the content has a uniform temperature which actually is not the case as shown in fig. 3. Taking the maximum temperature - like it was done in this example - is truly conservative, but leads to high pressures.

There are some lessons to be learned from the dependencies shown in the diagram in fig. 6:

a) The driving force for the pressure build-up is the temperature.
b) Keeping the filling degree low avoids high pressures. A filling degree of less than 0.1% results in a maximum pressure of 0.5 MPa (5 bar) even at a uniform temperature of the content of 800 °C. Two actions help to minimize the filling degree: At first the amount of water must be reduced to its minimum. And secondly, which is important, a huge free volume inside the package helps to reduce the maximum pressure, too. This leads to the following consideration: Thinking of the content as a ternary composite of solid, water and free volume, even with a fixed percentage of water the maximum achievable pressure depends on the ratio of solid and free volume. This fact leads to the necessity of an exact content description by the applicant. All possible compositions of the content must be taken into account for the determination of the maximum possible pressure build-up.

DISCUSSION OF THE RESULTS FROM BOTH APPROACHES

Both, the CFD/FE model and the steam table model, lead to a maximum pressure build-up in the same range. Due to the assumptions to be done and the uncertainties due to the lack of information and also due to disregarded processes (like vaporization and condensation) these results are not entirely reliable. It became clear that both approaches require an exact description of the cask and of the content to result in a best possible way. As the description of the cask is mostly accurate, the crucial point is the content description. It has to be stated that a comprehensive content description, which often represents a single state, is in general not comprehensive concerning the pressure build-up due to the thermal test.

The essential from the investigation of the CFD/FE model is that not only the maximum amount of content is the relevant state. Furthermore a partly filled (or nearly empty) package could be the worst case concerning the pressure build-up. The essential from the investigation of the steam table is that all ratios of the three phases (solid, liquid, gas) must be investigated to find the relevant composition of the content. Regarding all ratios of the three phases implies the requirement that all possible amounts of content are to be taken into account.

CONCLUSION

Two approaches for the determination of a pressure build-up due to vaporization caused by the thermal test were investigated. It became clear that the description of the content has to be as exact as possible to get the best possible results concerning the pressure build-up. It cannot be stated primarily if a package filled to its maximum, or a nearly empty package results in the relevant state regarding the pressure build-up. Additionally the investigations have shown that even small amounts of energy could lead to pressures inside the packages, which make it impossible to fulfill regulatory requirements.

Due to the inevitable uncertainties both approaches shown above must be seen as tools for an approximation of the actual values. Doing this kind of pressure build-up analysis first of all it is to be checked if all boundary conditions are conservative. Then every variation of possible contents must be analyzed, except of those which surely not represent the decisive case (e.g. just solids). Finally the results have to be checked for plausibility and assessed from the engineers point of view.

Doing these assessments either by computational calculation or by analytical calculation as shown above, three possible outcomes arise:

- The package design has a very high margin of safety. E.g. the package is covered totally by impact limiters. Regarding to the pressure build-up the package could fulfill the regulatory requirements.
- The package design could not fulfill the regulatory requirements, e.g. because of an amount of energy which leads to total vaporisation of all liquid inside the package.
- All assessments with results between those two extreme outcomes (comply/not comply) must be stated as unacceptable regarding the fulfilment of the regulatory requirements, because no trustworthy results regarding the pressure build-up inside the package could be achieved.

To get realistic and reliable results in safety assessments, experimental fire tests should be taken into consideration. Since in general experimental fire tests could represent just a single state of content, the requested content for a package should be specified exactly. This helps to minimize the number of experimental fire tests in a safety assessment. However, this paper shows the difficulties of assessing safety of packages containing wet content. From the authority assessment point of view, drying of the content could be an effective way to avoid the pressure build-up and the associated challenges for the safety assessment.

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