FIRE RISK ON HIGH-PRESSURE FULL COMPOSITE CYLINDERS FOR AUTOMOTIVE APPLICATIONS

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ABSTRACT

In the event of a fire, the TPRD (Thermally activated Pressure Relief Device) prevents the high-pressure full composite cylinder from bursting by detecting high temperatures and releasing the pressurized gas. The current safety performance of both the vessel and the TPRD is demonstrated by an engulfing bonfire test. However, there is no requirement concerning the effect of the TPRD release, which may produce a hazardous hydrogen flame due to the high flow-rate of the TPRD. It is necessary to understand better the behavior of an unprotected composite cylinder exposed to fire in order to design appropriate protection for it and to be able to reduce the length of any potential hydrogen flame. For that purpose, a test campaign was performed on a 36 L cylinder with a design pressure of 70 MPa. The time from fire exposure to the bursting of this cylinder (the burst delay) was measured. The influence of the fire type (partial or global) and the influence of the pressure in the cylinder during the exposure were studied. It was found that the TPRD orifice diameter should be significantly reduced compared to current practice.

1 - INTRODUCTION

Hydrogen is expected to be a valuable energy carrier for the 21st century. Its application in Fuel Cell Electric Vehicles is considered particularly important as they give the lowest carbon solution for medium/larger cars and longer trips [1]. Among the several existing hydrogen storage methods, high-pressure hydrogen storage is the most favorable for long term viability [2]. A fully wrapped composite cylinder is the most investigated option because of its lightness which allows the storage of a large volume of hydrogen at very high pressure. Type III cylinders (with a metal liner) are widely used for CNG (compressed natural gas) applications at 25 MPa and H_2 applications at 35 MPa, and type IV cylinders (with a plastic liner) tend to be the standard for 70 MPa H_2 applications as they are less susceptible to fatigue cracks.

The carbon-fiber/epoxy composite laminate is the load-bearing unit in the hydrogen storage vessel. The liner is only a few millimeters thick and only used for gas tightness. The composite laminate is sensitive to fire and high temperature which would degrade its mechanical properties. The safety strategy for a fully wrapped composite cylinder involved in a fire consists in preventing the cylinder from bursting by allowing the release of the hydrogen through a Thermal Pressure Release Device (TPRD), activated by a thermo-fusible material. Pressure-activated Relief Devices (PRD) are not used as the excessive pressures required for activation would not be achieved. Reviews of the accident literature on the CNG and H_2 composite cylinder [3, 4] showed that the cause of accidental burst of cylinders was mainly a localized fire or a wrong design of the size of the TPRD orifice.Then, overpressure and fragments from the burst cylinder could have catastrophic consequences.

Standard drafts (such as ISO/DIS 15869.3, CGH2R-12b, SAE J2579 [5]), regulatory drafts (such as CGH2R-12b), and regulations (e.g. [6]) define the testing of composite hydrogen storage vessels for fire impact. The test conditions are either global engulfing fire conditions or partial fire exposure. However, these tests are always performed on cylinders equipped with a TPRD so they cannot prove the intrinsic safety of the cylinders. Moreover, specific thermal protection covering the cylinder is rarely taken into account even if it would allow a longer activation delay for the TPRD and/or a longer emptying duration of the cylinder. Finally, the danger resulting from the TPRD flow rate and induced flame length is not taken into account. In order to protect people from both cylinder burst and hydrogen flame, it is necessary to optimize the fire protection of the cylinder, which may include a TPRD and/or a specific thermal protection cover. In that perspective, it seems necessary to understand better the behavior of an unprotected composite cylinder exposed to fire.

So far, the safety performance of a fully wrapped composite cylinder without a TPRD exposed to fire has rarely been studied. Weyandt [9] performed a test on an 88 L hydrogen type III cylinder pressurized to 31.8 MPa which was placed in a typical SUV (Sports Utility Vehicle). The cylinder burst after being exposed to the propane bonfire for 12 min 18 sec. The cylinder failed through the bottom, destroying the automobile and bonfire pan, and launching the remains of the cylinder 41 m from the test location. A blast wave pressure of 143 kPa was measured 1.2 m from the vehicle and 12 kPa was measured 15 m from the vehicle. Limited hazards would be expected below 3 kPa which was estimated to occur at approximately 25 m, based on extrapolation of the blast-pressure data. Weyandt [10] and Zalosh [11] also performed a test on a type IV 72.4 L composite vessel without a TPRD which was pressurized to 34.3 MPa and completely engulfed in a propane bonfire. The cylinder burst after 6 min 27 sec. Blast wave pressures measured along a line perpendicular to the cylinder axis were 18% to 25% lower than the values calculated from ideal blast wave correlations using a blast energy of 13.4 MJ, which is based on the ideal gas internal energy at the 35.7 MPa burst pressure. The resulting hydrogen fireball maximum diameter of 7.7 m was about 19% lower than the value predicted from existing correlations using the 1.64 kg hydrogen mass in the tank.

More tests have been performed on hydrogen cylinders equipped with a TPRD. Weyandt [10] compared different TPRD technologies and the influence of filling levels on six composite cylinders. Tests were performed on 35 MPa type III (diameter 410 mm length 970 mm) and type IV (diameter 410 mm length 840 mm) cylinders filled at 10%, 25% or 100%. The pressure relief valves of the cylinders activated between 1 and 2.5 min . In all tests, the cylinders content was released without cylinder burst. Weyandt concluded that the bonfire test was not sufficient to assess a cylinder's ability to withstand fire exposure. The test evaluates only whether the test setup can engulf a pressure relief device in flame but does not provide a safety measure on how long a cylinder can withstand a small fire/heating scenario that does not directly heat the pressure relief device nor on how long a cylinder can withstand a larger-sized fire/heating scenario should a pressure relief device be faulty or bypassed by a user. Suzuki [12] performed a real-world fire test on a 4-door Sedan vehicle type with two hydrogen 35 MPa type III cylinders using one glass-bulb type TPRD (110 °C activation) each. The fire was started in the ashtray in the front door of the car and the cylinders were located in the trunk of the vehicle. The TPRD activation time was between 14 min 36 sec and 17 min 4 sec but the flame started to heat the storage units only after 6 min to 8 min. Zheng [13] performed a bonfire test with natural gas on a 35 MPa type III cylinder (400 mm outer diameter and 900 mm length). The cylinder was equipped with a TPRD with a 6 mm orifice and filled with H_2 at 28.4 MPa. The TPRD opened after 6 min 17 sec with a cylinder pressure increase of 10% and the cylinder did not burst.

The objective of the present study is to understand better the intrinsic resistance of hydrogen fully-wrapped composite storage to thermal aggression (partial or global fire) in order to develop either a thermal protection cover and/or to improve the TPRD design. In a first step, the net flux received by a metallic cylinder in such a bonfire and the reproducibility of the thermal impact conditions are assessed. In a second step, experiments performed on a composite cylinder (type IV, epoxy resin and carbon fibers, 36 L, 70 MPa) are described. The influence of partial or global fire impact and the influence of the initial filling pressure on burst delay are studied. As a final step, it is confirmed that a controlled release with a small orifice diameter is sufficient to avoid burst phenomena.

2 - BONFIRE CHARACTERISATION

In order to characterize the pool fire used to perform the bonfire test required by current regulation and standards [5] [6], a steel cylinder sealed at both ends (diameter 330 mm and length 900 mm and 12 mm thickness) and filled with air was used to assess the heat flux received by the cylinder by monitoring the pressure increase inside the cylinder. The tests were performed at the INERIS facility in a gallery carved into the rock, 80 m long and 10 m² section (about 3.50 m wide and 3 meters high), equipped with forced air ventilation of 10 m^3/s.

The fuel selected was heptane, a hydrocarbon referenced for this type of experiment which allows good visibility. The fire was made from a rectangular pan of heptane (dimension 0.6 x 1.2 m^2). In order to reach constant flame behaviour and to limit the heptane quantity, the liquid heptane level in the pan was regulated by heptane injection on the basis of a thermocouple placed in the pan. It should be noted that during a preliminary test, the cylinder burst formed a large quantity of heptane mist which produced a violent secondary explosion. To allow visibility, it was necessary to extract fumes, but baffles were also necessary to protect the fire (Figure 1).

Figure 1: On the left: fire is growing (first phase) – on the right: stationary phase

The power developed by the fire can be estimated from the amount of fuel consumed during the test given the 100% combustion efficiency of heptane. The fuel consumption can be estimated from the filling rate of the tank (5.71 L / min) during the fire. This calculation gave a heptane consumption of 48 g/m²/s, and total power of 1.5 MW for a pan of 0.72 m².

The pressure on the small pipe connected to the cylinder was measured with a pressure sensor (Kistler piézorésistive 0-10 bar type 4045A10).

The temperature inside the cylinder was estimated assuming a perfect gas behaviour for heptane. The net flux received by the cylinder calculated from the air temperature increase inside the cylinder and the rise in temperature of its wall is given in Figure 2 for two tests. They are very similar: it confirms the reproducibility of the thermal load imposed by the bonfire on a cylinder and shows that the thermal flux is not constant with time.

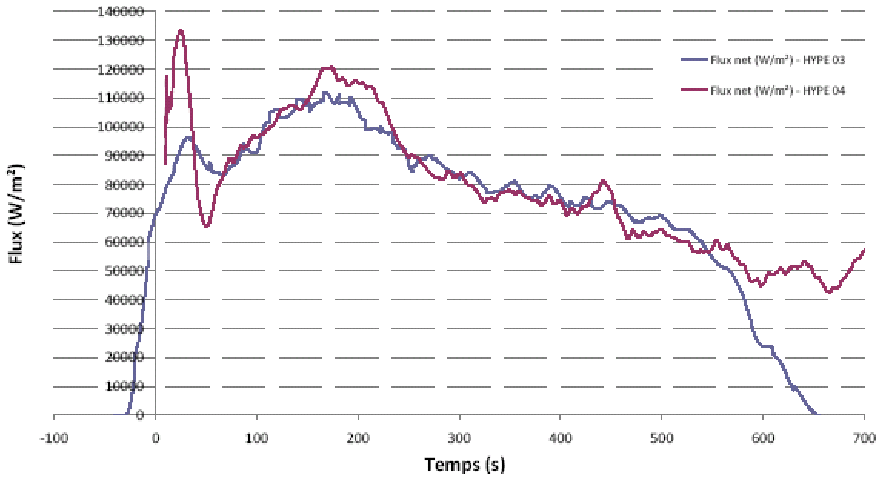


Figure 2: Net flux received by the cylinder for two preliminary tests

The first peak (in the first 50 seconds) can be explained by the low flames which brought convective heat to the lateral wall of the cylinder. After 200 seconds, the heat flux reached a maximum of 110 kW/m^2 to 120 kW/m^2. It corresponds to the time it took for the flames to completely cover the cylinder in the video analysis (Fig 1). This is to be compared to the 150 kW/m2 value given as a reasonable value for the heat flux at the flame surface [14]. After this second peak, the heat flux decreased as the outside wall temperature increased so that the convection flux decreased.

3 - BONFIRE TESTS ON COMPOSITE CYLINDER

Five tests were carried out on a fully-wrapped composite 70 MPa 36 L 34 kg type IV cylinder with a design coefficient of 3 (its bursting pressure is above 210 MPa). In order to better understand the influence of internal pressure on the intrinsic resistance time of a composite cylinder in a fire, 3 bonfires were performed without a TPRD and with initial pressures of 700, 350 and 175 bar of helium, respectively. Then, a partial fire test was performed, without a TPRD, with an initial pressure of 700 bar to assess the influence of the surface impacted by fire. Finally, a bonfire test was performed with a helium release simulating a TPRD (orifice diameter = 0.5mm) to check if such a small orifice and low pressure decrease could prevent the cylinder from bursting.

3.1 - Experimental set-up

The composite cylinders were pressurized with helium for safety reasons (hydrogen explosion in the gallery). One pressure sensor measures the pressure inside the cylinder (FGP 1000 bar type P101). Five thermocouples were fixed to the external surface of the cylinder with metallic loops, and another thermocouple was placed in the fire (50mm under the composite cylinder). The bonfire was made with heptane contained in a 0.8 x 1.2 m^2 pan. As for preliminary tests, the fire was stabilized with deflectors which were positioned around the fire and the gallery ventilation was on. The cylinders were placed 100 mm above the heptane surface.

Figure 3: Experimental apparatus for bonfire tests (on the left: global fire – on the right: partial fire)

For the partial fire test, only a half of the cylinder was placed above the pool (as shown in Fig. 3), the rest of the cylinder being protected by a thermal shield.

After the bonfire tests without a TPRD, a last test was performed to check if the cylinder could withstand a bonfire without bursting using a smaller release orifice than usual. A release device was designed and installed in the gallery to simulate the emptying of the cylinder through a PRD. For the purpose, an orifice with a diameter of 0.5 mm was used, which opened 90 s after the start of the fire. The time of 90 seconds was chosen to represent the maximum delay for a TPRD activation in a bonfire. This activation delay is a maximum value from Air Liquide experience of bonfire tests performed with TPRDs. It is also the maximum value allowed to comply with the American standard CGA S-1.1 [15] for TPRD activation at 600°C. However, it should be noted that when choosing a TPRD, it is critical to check its activation delay in a bonfire as it may be higher than 1.5 min as measured by Weyandt (10) and explained in the introduction.

The following table summarizes the conditions of the tests and the thermocouple (Type K – 1mm) positions on the cylinder (TC1-TC5) and 50 mm under the cylinder (TC6).

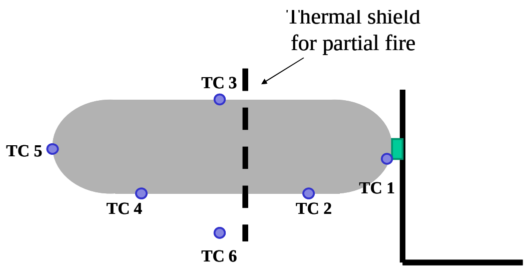
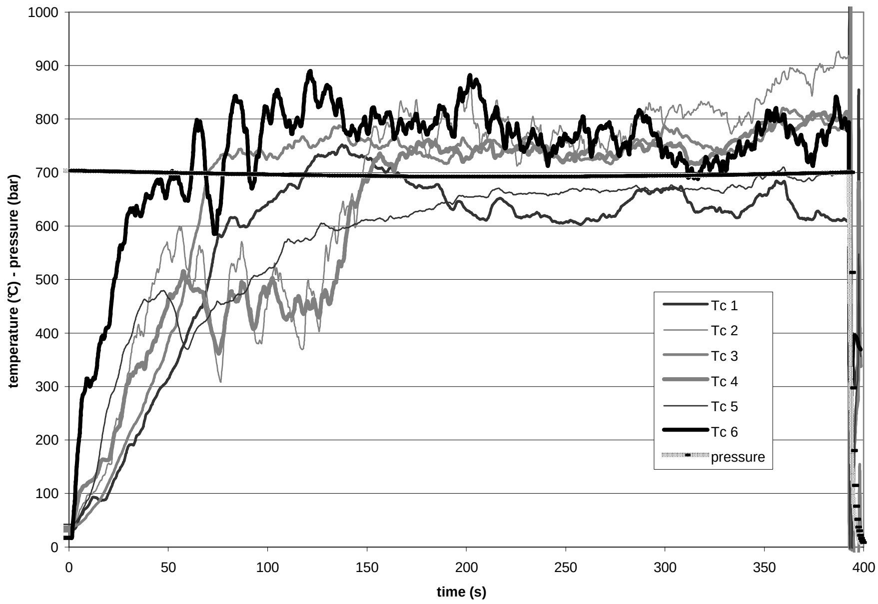
Cylinder type	Initial pressure	Fire configuration	TPRD	Thermocouple positions
type IV, 36L	175 bar	engulfed in fire	no	
	350 bar			
	700 bar			
	700 bar	partially in fire		
	700 bar	engulfed in fire	yes	

Table 4: Table summarizing the test conditions

3.2 - Results

The following figures show an example of the external temperature and inside pressure measurements for two of the tests.



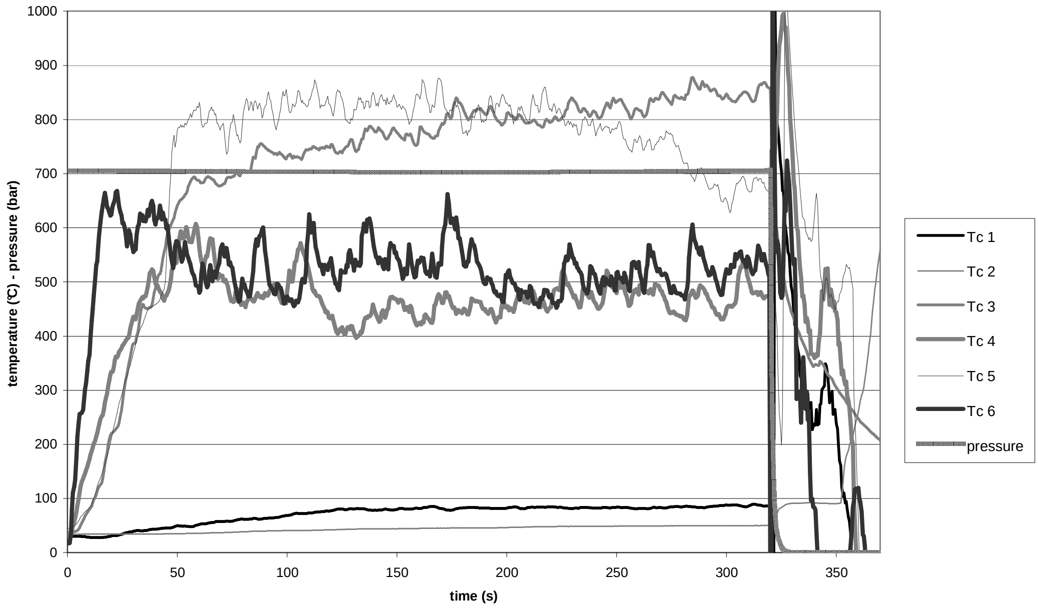


Figure 5: Temperature and pressure measurement for global bonfire test with initial pressure of 703 bar (top) and partial fire test with 706 bar initial pressure (bottom)

In all tests, the temperatures measured in the flame (TC6) were above 600°C after only a few seconds (maximum 40 seconds) except for the partial fire test where the TC6 thermocouple was placed in the immediate vicinity of the baffle, which reduced its temperature. The temperature of at least one thermocouple on the cylinder indicates a minimum temperature of 590 °C and is maintained for the remaining duration of the test as required in ISO_DIS15869 bonfire specification. However the 800°C required in SAE J2579 draft for engulfing conditions was not systematically reached.

Slight pressure drops could be found in early testing. They were due to the cooling (relaxation) of the gas after the filling procedure had been stopped. Cylinders are usually pressurized at pressures slightly above the required pressure to anticipate the drop in pressure due to cooling. This pressure drop does not always have the same amplitude (see min pressure ratio in table 6); it depends on the speed at which the cylinder is pressurized and on the initial pressure in the source container.

type of fire	initial pressure	burst pressure	time before burst	Pressure ratio extreme values : Min [P(t)/Pini] - Max[P(t)/Pini]
engulfed in fire	703 bar	703 bar	6 min 32s	0.99 – 1.00
partial	706 bar	706 bar	5 min 20s	0.98 – 1.00
engulfed in fire	356 bar	378 bar	9 min 49s	0.99 - 1.06
engulfed in fire	178 bar	No	no burst - leaks after 11 min 4s	1.00 - 1.125

Table 6: Table summarizing the results of the tests

There was no pressure increase inside the composite cylinder during the first 200 seconds after the start of the fire. This shows the significant thermal inertia of the composite material. The pressure increase before cylinder rupture or leak was either null or very low (The maximum pressure increase was +12.5% after 11 minutes in the bonfire for the cylinder whose initial pressure was 178 bar). Thus, before the opening of the release device, the pressure inside the cylinder was still the same.

The bursting delays are of the same order of magnitude as found by Weyandt [9, 10] (6 to 12 min for cylinders which were twice as large as the cylinder studied here, but with half of the design pressure).

3.2.1 – Influence of the fire engulfing conditions

Comparison of global bonfire and partial fire exposure results (Figure 7) shows a surprisingly larger resistance delay (+22%) for global fire than for partial fire exposure.

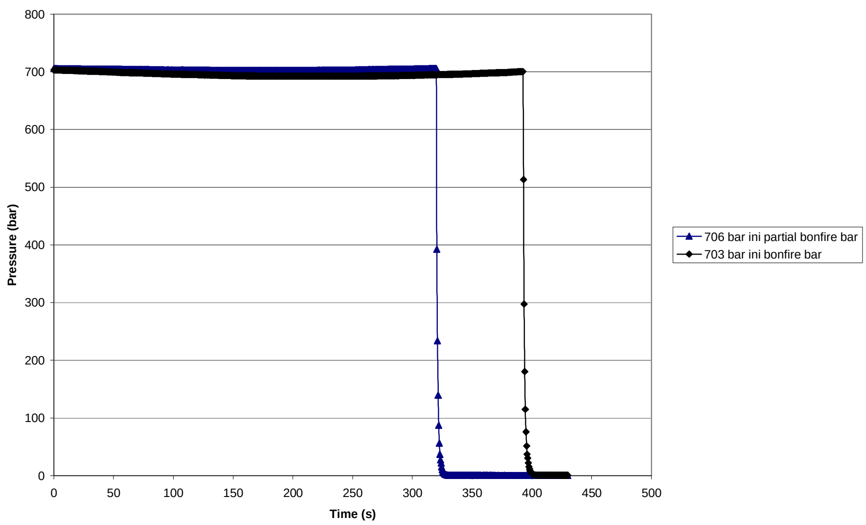


Figure 7: Comparison between bonfire versus partial fire pressure evolution

Maximum temperatures measured on the composite surface seem to be in the same order of magnitude (750 to 850 °C) This 22% increase in burst delay may be explained by a local heating effect which was not measured by the thermocouples. We would need additional tests results on this respect but we can conclude that the burst delay does not depend significantly on the size of the surface impacted by the fire even if this size is divided by a factor of 2.

3.2.2 – Influence of initial pressure

As shown on figure 8, the higher the initial pressure, the shorter the resistance time. However, the main result of this test series is that if the initial pressure of the storage is less than 178 bar, this composite storage unit leaks after 11 min and does not burst.

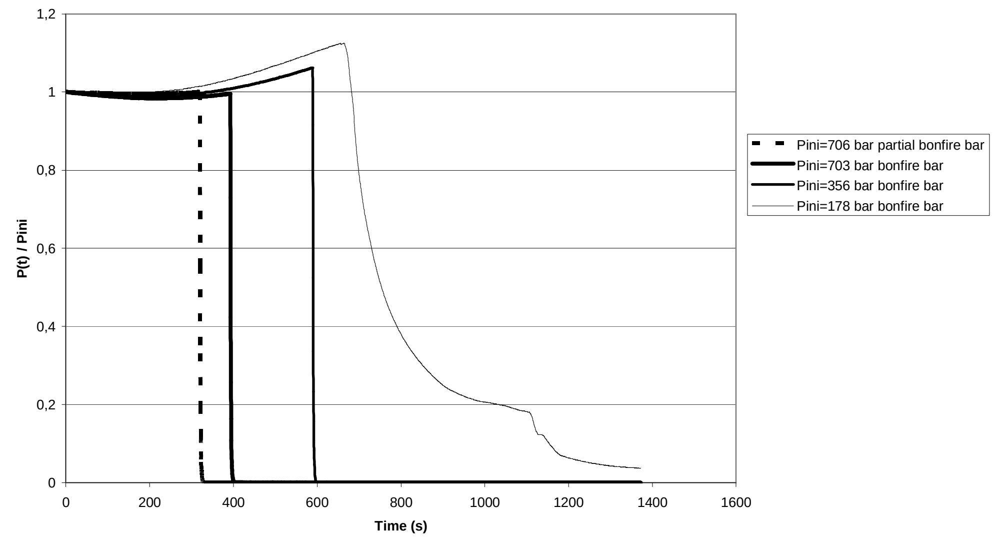


Figure 8: Pressure ratio ($P(t)/P_{ini}$) evolution as a function of time

The failure time (burst or leak) of the cylinder is multiplied by a factor of 2 between the two extreme filling pressure tests: 706 bar and 178 bar. Therefore, the initial pressure of the cylinder seems to have a greater influence on the burst time than the size of the impacted surface.

After the bonfire test, the cylinder with an initial pressure of 178 bar was tested to look for leakage areas using soap bubbles. The following pictures (Fig. 9) taken during the leak check, show that the cylinder was leaking across its entire surface with slightly more leakages at the ends.

Figure 9: Cylinder Pini=178 bar after bonfire test (left) and during leak test (right)

The epoxy resin seems to have disappeared but the carbon fibers did not burn. The sharp drop in temperatures recorded after 15 minutes, including the thermocouple positioned between the cylinder and the pan of heptane corresponds to when the fire was stopped. Some thermocouples showed a less rapid drop in temperature due to the fact that the resin continued to burn even after the fire stopped.

Assuming a sonic flow and considering helium as an ideal gas with an isentropic release, the pressure inside the cylinder emptying with an orifice surface A as a function of time is given by equation (1) [16]:

$$P(t) = P(t = t_1) \times Exp\left(-\frac{(t - t_1)}{C_1} \right)$$

$$C_1 = \frac{V}{A}\left(\frac{MW}{\gamma R T_0} \right)^{0.5} \left(\frac{2}{\gamma + 1} \right)^{-\frac{\gamma+1}{2(\gamma-1)}} \qquad (1)$$

Where P– Pressure, Pa, t - time , s , t_1-initial time, s, A- area of the orifice, m^2, V- volume of the cylinder , m^3, MW – molecular weight of the gas, g/mol, γ – specific heat ratio, -, R – universal gas constant , J/mol K, T_0 – stagnation temperature.

From equations (1), the equivalent leak diameter corresponding to the first pressure decrease slope when the cylinder started leaking was about 0.7 mm. This leak size was not constant in time. It increased significantly 7 min 30 sec after the cylinder started to leak (note a change in the slope in Fig. 8 at 1112 sec, then at 1135 sec and 1146 sec) even though the bonfire was stopped 4 minutes after the cylinder started to leak. The authors believe that this leak was due to the liner melting or that the liner was weakened by high temperatures and by the strength of the gas pressure: then the gas was released through the composite layers.

3.2.3 – Controlled and slow release

With the results of these three tests without PRD, one may define the minimum release conditions which would lead to leakage from the cylinder and no burst. Indeed, the 178 bar test gives a pressure/time threshold for the composite cylinder corresponding to the maximum pressure reached during the test (i.e. the pressure at which the leak occurred) and its corresponding time: in the case of the studied cylinder, this threshold can be estimated to 200 bar and 11 minutes. To avoid a burst, a TPRD should detect the high temperature and allow the release of the H_2 to decrease the cylinder pressure under 200 bar in less than 11 minutes.

From the above equations, we calculate that for a 36 L cylinder containing helium at a pressure of 700 bar, a release device with an orifice of 0.5 mm opening after 90 s will empty the cylinder quickly enough to decrease the pressure below 200 bar after 11 minutes. This size of orifice is to be compared to the significantly larger release diameters currently used for high pressure hydrogen cylinder TPRDs (3.6 mm to 6 mm). From the model proposed by W. Houf [17], we can extract the following correlation for a sonic release.

$$L_{flame} = 426.2 \times P^{0.459} \times d_{opening} \qquad (13)$$

Where L_{flame} – flame length, m, P – Pressure, MPa, $d_{opening}$ -orifice diameter, m

Considering the direct proportion between the opening diameter and the flame length [17], such a reduction would allow a decrease in flame length from 11 to 18 m to 1.5 m at 70 MPa.

Figure 10 compares the evolution of the pressure inside the cylinder during the gas release, from theoretical calculations and during the test.

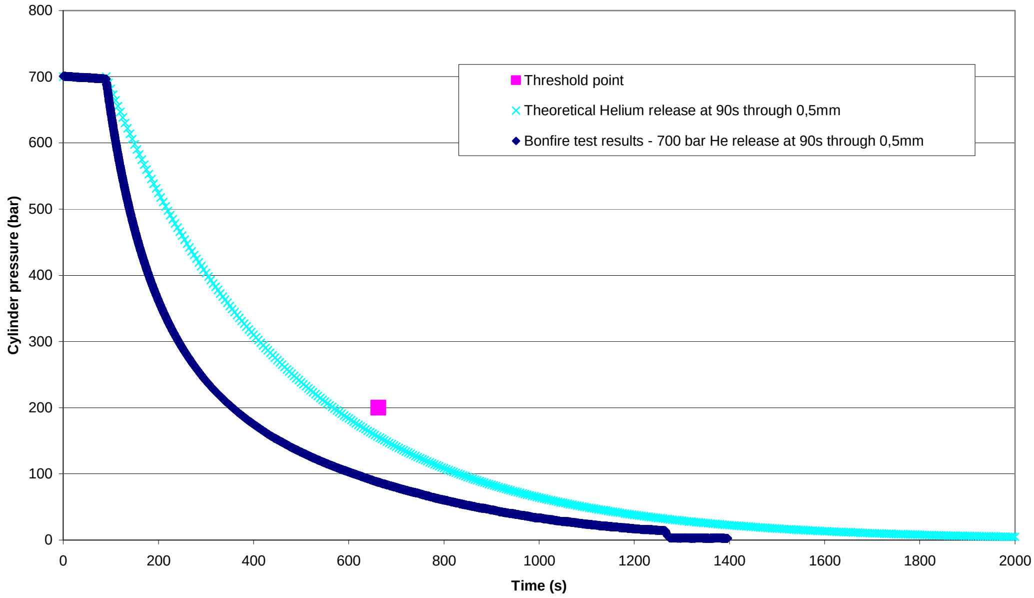


Figure 10: Theoretical versus measured cylinder pressure as a function of time during release (orifice diameter=0.5 mm)

The decrease in pressure measured in the bonfire test is faster than calculated with equation 1. Using Equation 1, the measured pressure release would correspond to a 0.6 mm orifice diameter. This may be due to the temperature decrease in the cylinder which is induced by depressurization and to the fact that the gas is considered ideal and the release isentropic. It may also be explained by the real orifice diameter which may be a little bit larger than the expected 0.5 mm.

This small orifice allowed the cylinder to leak instead of bursting as was intended. It allowed a reduction in the flame length by a factor 10. Similar results have also been obtained in a test campaign involving 2.4 L 70 MPa type IV cylinders, at different initial pressures [18]. For such a small cylinder it was found that an orifice diameter as low as 0.1 mm for a TPRD, opening within 2 minutes in a bonfire, could reduce the flame length to less than 0.5 m.

These results cannot be generalized to any fully-wrapped type IV composite cylinder. Many parameters (e.g. resin thermal properties, carbon fiber content, cylinder volume, pressure design, cylinder thickness and liner material) will have a strong influence on the intrinsic resistance of fully-wrapped type IV composite cylinders to bonfire. In addition, the design of an optimized TPRD orifice will not only be linked to the intrinsic resistance of the cylinder but also to its activation delay and to any thermal shield added to the cylinder.

4 - CONCLUSION

These experiments help to characterize the thermal impact of a bonfire and to understand the behavior of cylinders submitted to thermal stress (fire) and mechanical stress (internal pressure).

The tests performed without a release device showed that the resistance time of a composite cylinder is of the same order of magnitude for a localized fire (where only half of the cylinder was exposed to fire) and for a global bonfire, which are commonly performed for fire performance tests of composite cylinders according to ISO15869 or European Regulation ECE 79-10. The cylinder as a whole needs to be protected from localized fire impact as demonstrated in these experiments or use must be made of thermal protection for the cylinder itself.

For the tested Type IV 36 L cylinder, a pressure-time threshold was defined to predict whether the cylinder without a release system would burst or leak. Thus, a release can be modeled and the orifice diameter can be defined so that the pressure will fall below the critical pressure at the threshold time.

The orifice diameter is limited by the safety objective in terms of flame length. A compromise can be found to release the hydrogen quickly enough in order to decrease the pressure below the threshold pressure at the critical time but slow enough to limit the flame length. Depending on the storage volume, pressure design and composite thickness, thermal insulation may or may not be needed in order to allow a larger activation delay and venting time through the TPRD. Without any thermal protection, in the case of the studied cylinder, the orifice diameter could be decreased by a factor 10 compared to current practices, allowing the flame length and consequently the safety distance to be decreased by the same factor.

Regarding the development of a fully-wrapped composite type IV cylinder, we recommend performing similar tests to define the pressure and time threshold under which the cylinder leaks but does not burst. This definition would contribute to the design of a safer TPRD which would allow a smaller release flow-rate. Results from such tests will lead to improved cylinder designs, as the leak time and pressure will be linked to parameters such as the cylinder volume, the composite thickness and the liner material and design.

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