# PRD HYDROGEN RELEASE AND DISPERSION, A COMPARISON OF CFD RESULTS OBTAINED FROM USING IDEAL AND REAL GAS LAW PROPERTIES

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## ABSTRACT

In this paper, CFD techniques were applied to the simulations of hydrogen release from a 400-bar tank to ambient through a Pressure Relieve Device (PRD) 6 mm (¼") opening. The numerical simulations using the TOPAZ software developed by Sandia National Laboratory addressed the changes of pressure, density and flow rate variations at the leak orifice during release while the PHOENICS software package predicted extents of various hydrogen concentration envelopes as well as the velocities of gas mixture for the dispersion in the domain. The Abel-Noble equation of state (AN-EOS) was incorporated into the CFD model, implemented through the TOPAZ and PHOENICS software, to accurately predict the real gas properties for hydrogen release and dispersion under high pressures. The numerical results were compared with those obtained from using the ideal gas law and it was found that the ideal gas law overestimates the hydrogen mass release rates by up to 35% during the first 25 seconds of release. Based on the findings, the authors recommend that a real gas equation of state be used for CFD predictions of high-pressure PRD releases.

## 1. 0 INTRODUCTION

The study of compressed hydrogen releases from high-pressure storage systems has practical application at the early stage of introduction of hydrogen and fuel cell technologies. Several demonstration projects for transportation and power markets use high-pressure stationary storage as storage media for hydrogen fuel. Understanding hydrogen behavior during and after the unintended release from a high-pressure storage device is important for development of installation codes and risk mitigation requirements (e.g. clearance distances and/or protective barriers). High pressure hydrogen releases may happen either due to accidental damage to a storage tank or connecting piping, or direct PRD releases. In the case where a storage tank is damaged to a large extent, a possible scenario is a relatively large opening to the environment leading to a fast release. The opening could be a crack or a hole in the vessel wall. The corresponding hydrogen mass outflows have to be regarded as variable and non-linear, since the conditions in the tank are changing quickly and both the stagnation pressure and the hydrogen density are decreasing accordingly with time. In general, the release flow of compressible hydrogen is critical (choked) at the leak orifice, which means that the upstream pressure is high enough for the release velocity of hydrogen to reach the speed of sound, or the maximum flow velocity possible, in the gas mixture. For a given constant downstream pressure, namely, standard atmospheric pressure, the decrease of the upstream stagnation pressure will reduce the mass flux from the sonic (choked) release to subsonic releases, in which the release velocity is below the local sonic speed for the gas mixture. It is a challenge to measure accurately experimentally hydrogen concentration cloud envelopes caused by the dispersions that result from the non-linear transient high-pressure hydrogen release from a storage tank.

In addition to experiments, computational fluid dynamics (CFD) simulations of hydrogen releases and dispersion were demonstrated to be an inexpensive approach to quantitatively predict the hydrogen

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dispersion in real time. The hydrogen releases and dispersion depend on the physical properties of hydrogen, the storage conditions (initial pressure, temperature), and, possible subsequent mechanical and physical interaction with the environment (wind velocity, ambient temperature). Literature search shows that there are CFD publications [1] related to the research in high-pressure hydrogen leaks and in variable mass release rates, but most of them exploit hydrogen properties expressed by the ideal gas law, which is quite different from hydrogen release behaviour at high pressures. Limited papers were found relevant to the hydrogen releases and dispersion by using real gas hydrogen properties. Among those applications of CFD methods using real gas relations, Venetsanos et. al. [2] simulated the hydrogen release rate, the resulting concentration volume and combustion of hydrogen in an actual hydrogen explosion, which took place in a built up area of central Stockholm, Sweden. The simulation results are consistent with the reported real situations.

Using the similar approaches, this paper presents a CFD technique incorporating both ideal and real gas laws to simulate a direct release from a crack or a hole on the vessel wall of a high-pressure storage system to ambient through a Pressure Relieve Device (PRD) 6 mm (¼”) round opening orifice. The objective of the current task is to investigate the extents of lower flammability limit (LFL) and 50% of LFL hydrogen clouds during PRD release from a hydrogen storage tank.

The TOPAZ network flow code [3], developed by Sandia National Laboratory, was used to address the changes of pressure, density and flow rate variations at the leak orifice during release while the PHOENICS software package [4], developed by CHAM Limited, predicted extents of various hydrogen concentration envelopes as well as the velocities of gas mixture for the dispersion in the domain. For a limited range of conditions, the Abel-Noble Equation of State (AN-EOS) was proved to be an accurate model for predicting the hydrogen real gas properties under high pressures [5], and was incorporated into the CFD model, implemented through the TOPAZ and PHOENICS software, to accurately simulate the hydrogen release and dispersion. The numerical results are compared with those obtained from using the ideal gas law.

## 2.0 MODELING SCENARIO DESCRIPTIONS

The release conditions for the two scenarios are:

| | |
|---|---|
| Tank: - 205 Litre Water Volume; | Pressure: 40 MPa (400 bars); |
| Ideal gas quantity of hydrogen in the tank: 5 kg; | PRD hole diameter: 6 mm (¼”); |
| Leak direction: horizontal; | Tank elevation above ground: 0.5 m. |

The PRD leak location is at the centre point of rounded end. As per the recommendation by IEC 60079-10 (for electrical classification of hazardous locations), it is assumed that there is a horizontal wind with a velocity of 0.5 m/sec in the direction of the leak. The ambient temperature is 20°C. The above phenomenon is classified as compressible release with variable flow rate. It should be noted that inside the tank, the conditions were assumed to be isentropic, while outside the tank – isothermal. Only choked part of the release was simulated.

## 3.0 MATHEMATICAL FORMULATIONS AND SIMULATION RESULTS

Results quoted below were obtained using a general CFD approach, where all the important physical processes were accounted for. Hydrogen convection, diffusion, buoyancy and transience were modeled based on the 3-D compressible Navier-Stokes equations and hydrogen mass conservation equation with the proper initial and boundary conditions. The compressible model only assumed that the temperature gradients were small in the domain and thus the gases (hydrogen and air) had compressible properties: the mixture gas density was a function of hydrogen mass concentration, temperature and pressure at the specified conditions.

The complicated 3-D hydrogen dispersion, which followed a high pressure choked release, was fully controlled by the turbulent flow. PHOENICS software was chosen for the purpose of modeling of the dispersion because it contains a number of validated turbulence models that allow for modeling of complex flow conditions. The LVEL model was selected as a proper turbulence model for computational tasks herein. The LVEL model allows for both laminar and turbulent flow conditions to be considered within one model. The LVEL subroutine computes local Reynolds numbers in every cell of the computational mesh and applies the local effective viscosity based on this number. The effective viscosity includes both laminar and turbulent components. This allows for accurate modeling of fluid flow conditions within the whole domain.

Another important feature of the modeling approach was the use of transient conditions for computing the releases and dispersion of hydrogen clouds, accounting for the transient behaviour of all calculated variables (pressure, gas density, velocity and hydrogen concentration) and the movement of hydrogen clouds with time.

To account for the effect of hydrogen buoyancy, the density difference model implemented in the PHOENICS was used. The dispersed hydrogen was driven by the buoyancy force caused by the density difference between the local mixed gas density and the standard reference air density.

### 3.1. Abel-Nobel equation of state

Under high pressure, hydrogen displays gas properties different from the ideal gas law predictions. For an ambient temperature of 293.15K and a pressure of 400 bars, the hydrogen density is about 25% lower than that predicted by the ideal gas law. The CFD real gas model used thermodynamic relations derived based on the AN-EOS, an equation of state, in which the hydrogen compressibility, $z_{H_2}$, is explicitly given in terms of empirical hydrogen co-density, $d_{H_2}$ [5]:

$$z_{H_2} = \frac{P}{r R_{H_2} T} = (1 - \frac{r}{d_{H_2}})^{-1} , \tag{1}$$

where ? , $P$, $T$ and $R_{H2}$ are the compressed hydrogen density, pressure, temperature and gas constant, respectively. Note that the hydrogen compressibility, $z_{H_2}$, is equal to 1 for the ideal gas law. The hydrogen gas constant, $R_{H_2}$, is 4124 J/(kgK). The hydrogen co-density, $d_{H2}$, is about 0.0645 mol/cm$^3$, or 129 kg/m$^3$. Equation 1 can be simplified as:

$$z_{H_2} = 1 + \frac{P}{d_{H_2} R_{H_2} T} . \tag{2}$$

The AN-EOS accounts for the finite volume occupied by the gas molecules, but it neglects the effects of intermolecular attraction or cohesion forces. Figure 1 shows the comparison of hydrogen compressibility using AN-EOS and the NIST (National Institute of Standards and Technology) data [6].

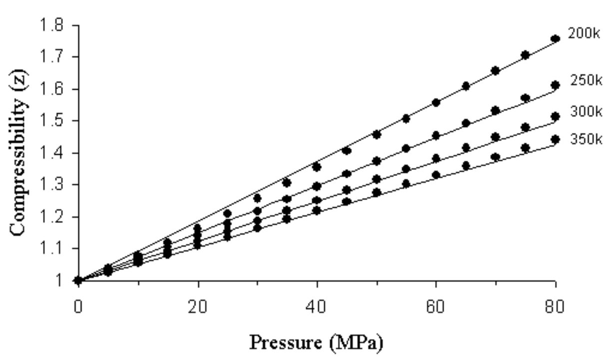


Figure 1. Hydrogen compressibility as a function of pressure.

The dots mark the NIST data and the lines mark the empirical data using AN-EOS. It is seen that the model accurately predicts the high-pressure hydrogen compressibility. At 400 bars, the real hydrogen gas model deviates from the ideal gas law ($z_{H_2}$=1) by 20% to 40% for temperatures from 200K to 350K.

## 3.2. Release rate

### *3.2.1. Ideal gas law correlations*

The hydrogen release rate was modelled by assuming a choked release (or sonic release) flow from the tank into the ¼" orif ice.

For an ideal gas the equation is of the form

$$\dot{m}_0 = C_d A \sqrt{r_0 P_0 g \left(\frac{2}{g+1}\right)^{\frac{g+1}{g-1}}} , \qquad (3)$$

where $\dot{m}_0$, $r_0$ and $P_0$ are the hydrogen mass flow rate, the gas density in the tank and the gas pressure in the tank, respectively, at *t=0*; *A* is the leak orifice cross-sectional area and $g$ is the ratio of specific heats for hydrogen. For hydrogen, $g = {C_P}/{C_V} = 1.41$, with $C_P$ and $C_V$ being the specific heat at constant pressure and constant volume, respectively. $C_d$ is the discharge coefficient. In this paper, $C_d$ was *0.95*, as recommended by Beek [1] for a rounded orifice with small contraction. The initial mass release rate corresponding to the tank with a pressure of 400 bars and a ¼" leak orifice is about $\dot{m}_0 = 0.753$ *kg/s*, based on the calculations from the ideal gas law.

Using the ideal gas law and solving the first-order ordinary differential equation for density with time, $r(t)$, by assuming a critical temperature at the leak orifice, the time-dependent release rate can be approximated as:

$$\dot{m}(t) = -V\frac{dr}{dt} = r(t)u(t)A \approx \dot{m}_0 e^{-\frac{C_d A}{V} t \sqrt{g\left(\frac{2}{g+1}\right)^{\frac{g+1}{g-1}} RT}} \approx 0.753e^{-0.110764t} , \qquad (4)$$

where *u(t)* is the hydrogen velocity at the leak orifice and *V* is the tank volume. The choked release lasts until the ratio of the pressure in the tank over the ambient pressure, namely, ${P_0}/{P_{atm}}$ is greater than or equal to $\left(\frac{g+1}{2}\right)^{\frac{g}{y-1}}$. It is estimated that the choked release lasts for about 47.7 seconds. Note that this approximation is only valid for the ideal gas model. The above ideal gas results were then used as the initial and boundary conditions for the dispersion simulation using the ideal gas law properties for the comparison to those from the real gas law.

### *3.2.2. Real gas law calculations*

Note that Equations 3 and 4 using ideal gas law are inappropriate for predicting a real gas release rate under high pressure. For a real gas release, a numerical simulation using TOPAZ with the implementation of AN-EOS was performed instead of the above correlations. TOPAZ is a transient one- dimensional pipe flow analyzer code for modeling the heat transfer, fluid mechanics, and thermodynamics of multi-species gas transfer in arbitrary arrangements of pipes, valves, vessels, and flow branches with the features of fully compressible model. The TOPAZ simulation showed that the mass release rate is lower than that from the ideal gas model. Figure 2 shows the comparison of the mass release rates obtained by Equation 4 using the ideal gas law and by TOPAZ using the real gas law. It can be seen that the mass release rate using real gas properties deviates from the ideal gas law by as much as 30% (at about 10 seconds). The ideal gas law significantly overestimates the hydrogen mass release rates in the first 25 seconds. The choked portion of the release, estimated by using the real gas law, lasts about 50 seconds in comparison to 47.7 seconds when using the ideal gas properties. The real gas law incorporated through AE-EOS is recommended for the hydrogen simulations when the experimental data is unavailable.

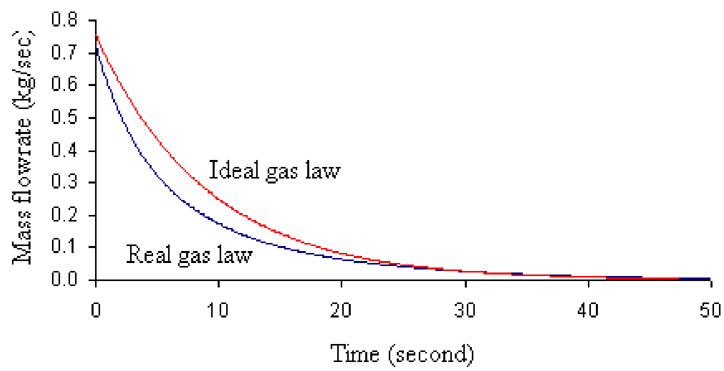


Figure 2. Choked hydrogen release rate.

Initial tank pressure: 400 bars. The leak orifice: ¼”. Black line: the real gas law using AN-EOS. The corresponding choked portion of the release lasts 50 seconds. Red line: the ideal gas law using $C_d = 0.95$. The corresponding choked portion of the release lasts 47.7 seconds. The ideal gas law overestimates the hydrogen release rate by up to 35% in the first 25 seconds.

### 3.3. Hydrogen dispersion

Using the two release rates shown in Figure 2, the CFD modeling of hydrogen dispersion was done separately for real and ideal gas laws as follows:

- An ideal gas dispersion simulation using the ideal gas law implemented through PLANT (a FORTRAN code generator in PHOENICS) exploited the ideal gas release rate obtained by Equation 4 as initial and boundary conditions at the orifice;

- A real gas dispersion simulation using AE-EOS implemented through PLANT exploited the real gas release rate obtained by TOPAZ as initial and boundary conditions at the orifice.

The compressed hydrogen releases to a domain of 40 m × 20 m × 10 m, which is assumed to be large enough for neglecting the boundary effects. To save the computational time, a structured grid of 35×18×21 for the half domain (40 m × 10 m × 10 m) was used for the symmetric hydrogen dispersion problem. Grid sensitivity study showed that this grid density yields sufficiently accurate numerical solutions.

Since the airflow caused by the forced convection from the 0.5 m/s wind can affect the hydrogen cloud introduced by the choked release, it is necessary to simulate the steady-state airflow first before trying to simulate the transient dispersion behaviour of the hydrogen cloud. A two-stage simulation was as follows: steady-state no-leak (before the release) simulation and the following transient hydrogen release simulation until the stop of the choked leak (the choked release duration). The velocity and pressure profiles obtained from the steady-state simulation (so called “before the release” simulation) were then used as the initial conditions for the choked release simulations, which were performed with a hydrogen leak at the specified rate and time increments according to the ideal and real gas laws, as shown in Figure 2.

During the simulations, the ground was assumed to have a no-slip boundary condition so the velocity was small close to the ground. The CFD modeling of the choked release was performed for 50 seconds by using the real gas AE-EOS and for 47.7 seconds by using the ideal gas correlation (Equation 4). The 3-D hydrogen cloud was investigated for the concentrations between 50% of LFL and LFL. Figure 3 shows the numerical results for the hydrogen concentration distributions (50% of LFL to 200% of LFL) 3 and 5 seconds after the onset of the release for both ideal and real gas laws. Sufficient accuracy in the calculations was guaranteed during the transient iterations by controlling the residuals for momentum, mass and concentration balance equations at each time step.

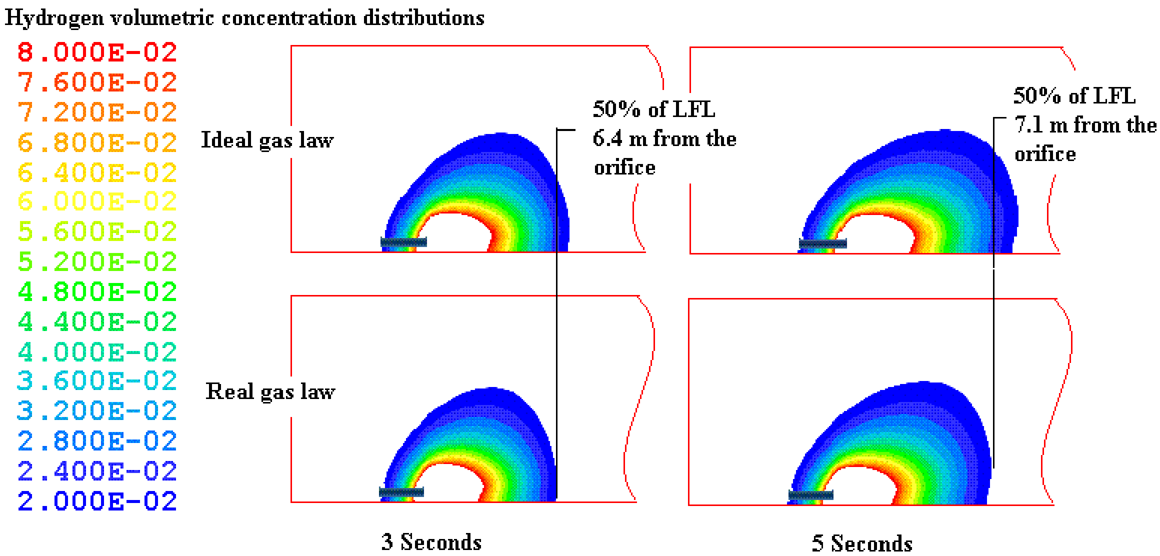


Figure 3. $H_2$ concentrations 3, and 5 seconds after the onset of the leak.

Top: Ideal gas law. Bottom: Real gas law. The label on the left marks the hydrogen concentration distribution range: from 2% to 8% (namely, 50% of LFL to 200% of LFL). The legends mark the distance between the leak orifice to the extent of 50% of LFL hydrogen cloud. The hydrogen cloud obtained from the ideal gas law is larger than that from the real gas law during the beginning of the release (See from the figures for the concentration distributions at 3 to 5 seconds).

Further investigation of the hydrogen cloud corresponding to 50% of LFL (2% vol.) shows that the cloud at 3 second has a horizontal extent of 6.84 m with using the ideal gas law and 6.41 m with using the real gas law. Table 1 shows the hydrogen cloud extent corresponding to 50% of LFL, LFL and 200% of LFL in the horizontal and vertical directions, 3, 5, 20 and 40 seconds after the onset of the leak. It is seen that at the end of 5 seconds, the hydrogen cloud extents obtained from the ideal gas law are more than 10% larger than those from the real gas law for different concentrations, translating to about 20% to 30% difference in hydrogen cloud volumes. This phenomenon can be explained by the fact that the ideal gas release rate is about 25% more than the real gas release rate at 5 seconds. At 20 seconds and after, the hydrogen clouds obtained using ideal gas law and real gas law have similar sizes as shown in Table 1 below.

Table 1. $H_2$ cloud extents at 3, 5, 20 and 40 seconds after the onset of the release

| Time | Concentration | Ideal gas law | | Real gas law | |
|---|---|---|---|---|---|
| | | Horizontal | Vertical | Horizontal | Vertical |
| 3 seconds | 200% of LFL | 3.11 m | 1.37 m | 2.82 m | 1.35 m |
| | LFL | 5.04 m | 2.60 m | 4.65 m | 2.48 m |
| | 50% of LFL | 6.84 m | 5.12 m | 6.41 m | 4.85 m |
| 5 seconds | 200% of LFL | 2.95 m | 1.37 m | 2.82 m | 1.25 m |
| | LFL | 5.11 m | 2.61 m | 3.80 m | 2.35 m |
| | 50% of LFL | 7.82 m | 5.40 m | 7.11 m | 5.11 m |
| 20 seconds | 200% of LFL | 3.35 m | 0.79 m | 3.34 m | 0.78 m |
| | LFL | 6.45 m | 2.07 m | 6.40 m | 2.02 m |
| | 50% of LFL | 11.85 m | 5.05 m | 11.80 m | 5.03 m |
| 40 seconds | 200% of LFL | 3.21 m | 0.78 m | 3.20 m | 0.78 m |
| | LFL | 6.01 m | 2.12 m | 6.02 m | 2.12 m |
| | 50% of LFL | 11.77 m | 6.00 m | 11.74 m | 6.10 m |

## 4.0 CONCLUSIONS

In this paper, a direct release of hydrogen from a tank with a high pressure of 400 bars to ambient through a PRD ¼" opening were simulated using the real and ideal gas laws. The Abel-Noble equation of state was incorporated into the CFD model to predict the real gas properties for hydrogen. It was estimated that it takes about 47.7 seconds to finish the choked portion of the release if the ideal gas law is used. The more accurate TOPAZ thermodynamic network codes using the real gas law predicts a 50-second choked portion of the release duration. CFD modeling of hydrogen dispersion was completed in two stages: a steady-state simulation before the onset of the leak followed by a transient simulation of the choked portion of the release. For the choked flow rate, the ideal gas law overestimates the hydrogen mass release rates in the first 25 seconds by up to 35%. This results in a longer horizontal extent and a larger volume hydrogen clouds when using the ideal gas law in comparison with the real gas law. This in turn leads to an unnecessary increase in clearance distances and sizes of hazardous zones associated with high-pressure releases of hydrogen. The authors predict that the differences between the ideal gas law and the real gas law results will increase at higher pressures. Based on the comparison of numerical results obtained from the two gas laws, the authors recommend that the CFD techniques incorporating the real gas law be used to evaluate hydrogen safety for high-pressure storage releases instead of using the ideal gas law.

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