EXPERIMENTAL INVESTIGATION OF NONIDEALITY AND NONADIABATIC EFFECTS UNDER HIGH PRESSURE RELEASES

Kuznetsov M.*[1], Pariset S. [1], Friedrich A. [2], Stern G. [2], Jordan T. [1]
[1] Karlsruhe Institute of Technology, Karlsruhe, Germany
[2] 2Pro-Science GmbH, Germany

**Corresponding author: kuznetsov@kit.edu*

ABSTRACT

Due to the nonideality of a high pressure hydrogen release the possibility of a two-phase flow and its effect on the dynamics of the discharge process was experimentally investigated. A small-scale facility was designed and constructed to simulate the transient blow-down of a cryogenic fluid through a small break. Gaseous and liquid nitrogen were planned to were used as a surrogate for GH2 and LH2. The results will complement the quasi-stationary safety regulation tests and will provide time-dependent data for verification of the theoretical models. Different orifice sizes (0.5, 1, 2, 4 mm) and initial N2 pressures (30 – 200 bar) were used in the tests. The measured time-dependent data for vessel discharge pressure, thrust, discharge mass flow rate, and gas temperatures were compared against a theoretical model for high pressure nitrogen release. This verification for nitrogen also assures the equation of state for hydrogen, which is based on the same methodology.

1.0 INTRODUCTION

The rapidly growing worldwide energy needs and the diversification of the energy mixed with an increasing share of renewable energies are challenging for existing energy infrastructures. More flexible solutions are required, especially those that integrate renewable energies more efficiently. With icefuel® (integrated cable energy system for fuel and power) a flexible system for energy storage, distribution and then reconversion has been developed [1]. The concept of icefuel® project was based on a grid of super-insulated cable for the transport of cryogenic fuels like liquid or supercritical pressurized hydrogen or natural gas. These cryogenic fuels serve as energy carrier as well as energy storage media.

A damage of such a cable with a small leak of hydrogen may lead to formation of hydrogen jet, which can be ignited being premixed with air [2]. Different flame propagation regimes may develop in such hydrogen – air jet depending on initial pressure and temperature in bulk volume (as icefuel® cable, for instance). The problem of safety analysis in this case is non-ideal state of the released gas and the gas inside the bulk vessel. This may result in difficulties to evaluate main properties of hydrogen jet structure during the release. One can see on the diagram of state of real hydrogen taken from [3] that two-phase flow of hydrogen is very probable in the case of pressurized supercritical hydrogen release starting from the conditions close to the operation of the icefuel® cable (see Fig. 1). Real side-view of two-phase flow of hydrogen for the test conditions #4006 (1- mm nozzle, 27 bar, 40K, see Fig.1) is shown in Fig. 2.

The dynamics of discharge mass flow rate may be calculated analytically from the integral form of the solution of Bernoulli equation written as follows

$$\dot{m} = C_D \cdot A_2 \cdot \max\left\{ \rho_2 \left[-2\left(\frac{p_1}{\rho_1}\right) \int_1^{\frac{p_2}{p_1}} \left(\frac{\rho_2}{\rho_1}\right)^{-1} d\left(\frac{p_2}{p_1}\right) \right]^{\frac{1}{2}} \right\}, \quad (1)$$

where C_D is the discharge coefficient; A_2 is the break area; p and ρ is the pressure and the density of the gas upstream (sub-index 1) and downstream of the flow (sub-index 2):

$$\dot{m} = C_D A_2 \begin{cases} \left\{ \gamma_u p_u \rho_u \left(\dfrac{2}{\gamma_u + 1} \right)^{\frac{\gamma_u + 1}{\gamma_u - 1}} \right\}^{\frac{1}{2}}; & \dfrac{p_d}{p_u} \le \left(\dfrac{2}{\gamma_u + 1} \right)^{\frac{\gamma_u}{\gamma_u - 1}} \\ \left\{ p_u \rho_u \dfrac{2\gamma_u}{\gamma_u - 1} \left[\left(\dfrac{p_d}{p_u} \right)^{\frac{2}{\gamma_u}} - \left(\dfrac{p_d}{p_u} \right)^{\frac{\gamma_u + 1}{\gamma_u}} \right] \right\}^{\frac{1}{2}}; & \dfrac{p_d}{p_u} > \left(\dfrac{2}{\gamma_u + 1} \right)^{\frac{\gamma_u}{\gamma_u - 1}} \end{cases}, \quad (2)$$

for critical (chocked) or subcritical flow under isentropic conditions.

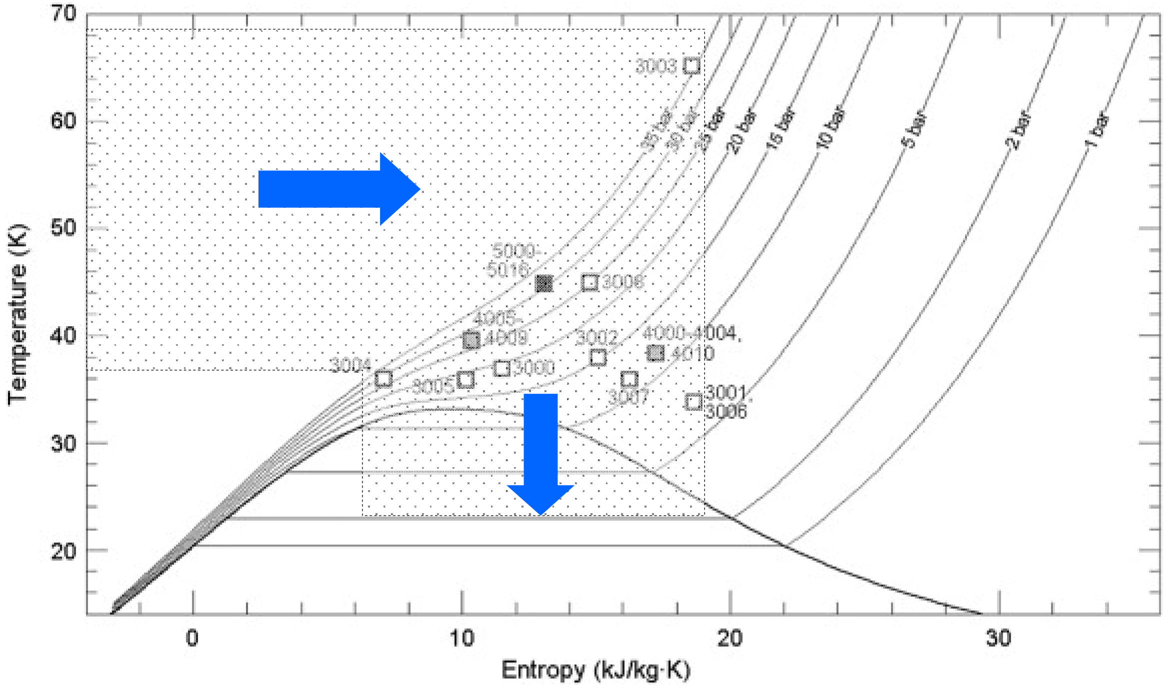

Figure 1. Temperature – entropy (T-S) – diagram of state of real para-hydrogen taken from [3]. Dashed lines show a range of the real gas state during the isentropic release process. Initial state of the gas (pressure and temperature) is shown as squared points with labelled test numbers.

Another problem of the high pressure hydrogen release is non-adiabatic nature of the process. Depending on initial pressure, diameter of the orifice and thermal isolation of high pressure volume the release conditions might be more close to iso-thermic or transient. The discharge coefficient takes into account the difference between theoretical and experimental mass flow rate:

$$C_D = \frac{\dot{m}_{\exp}}{\dot{m}_{theor}}, \quad (3)$$

where theoretical mass flow rate can be calculated analytically as for ideal gas by Eq. (2) or more precisely by numerical integration of Eq. (1). But even with the numerical integration, it is not clear what is the real pathway of the discharge process with respect to the constant entropy or real state of the gas, especially in the case of two-phase flow.

Objectives

The objective of current work is to obtain detailed experimental data on the high-pressure releases in wide range of initial pressures and nozzle diameters to take into account nonideality of the process. In order to simplify the conditions for two-phase flow, nitrogen will be used instead of the pressurized hydrogen. With these work, a capability of numerical and theoretical models for high pressure hydrogen releases will be validated against time-dependent experimental data.

Figure 2. Side view of cryogenic hydrogen jet [3].
Initial conditions correspond to the test #4006 (1- mm nozzle, 27 bar, 40K).

2.0 DIAGRAM OF STATE OF REAL GASES

Figure 1 shows the temperature – entropy (T-S) – diagram of state of real para-hydrogen. A T-S diagram is the most frequently used to analyze adiabatic high pressure gas releases. The thick solid line corresponds to saturation process of gas or liquid to two-phase gas-liquid state. This diagram allows to visualize the work done by or on the system and the heat added to or removed from the system. The heat transferred to or from a system equals to the area under the T-S curve of the process. Another advantage of T-S – diagram is the possibility to evaluate the ratio gas/liquid within two-phase area. The more left final state of the system appears within two-phase area, the more liquid phase is present in the system. As one can see from Fig. 1, practically in all experimental cases the two-phase state of the released substance might be reached in the case of isentropic discharge process (shown by blue arrow). The mist particles of liquid hydrogen could be clearly seen in Fig. 2 in case of pressurized cryogenic hydrogen release in air.

The problem for the experiments is that there is some difficulty to provide detailed measurements of pressure, temperature and mass flow rate for such pressurized cryogenic hydrogen systems because, for instance, initial bulk temperature hydrogen vessel must be below the ambient one to reach two-phase flow. This is the reason to substitute hydrogen by nitrogen at ambient temperature. Another reason to use inert nitrogen instead of hydrogen is to eliminate the possibility for hydrogen combustion as in the case of hydrogen release in air.

A T-S – diagram of state for real nitrogen was calculated using real gas equation of state from NIST database [4]. The T-S – diagram of nitrogen (Fig. 3) shows that starting from the pressure of 100 bar at ambient temperature the two-phase state could be reached during the isentropic process of high-pressure discharge to atmospheric pressure (vertical arrows). The ratio of liquid to vapour will increase with an initial pressure increase above 100 bar.

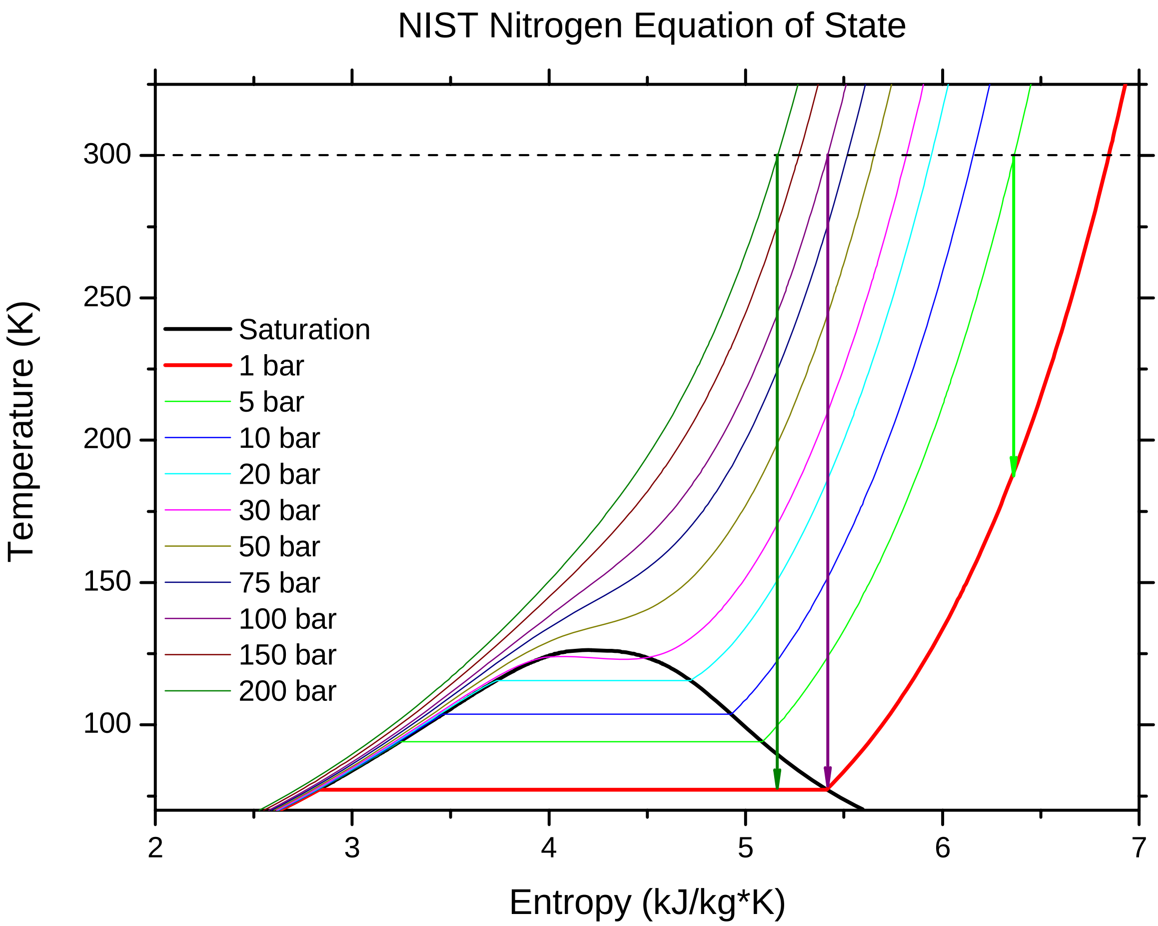

Figure 3. Temperature – entropy (T-S) – diagram of state of real nitrogen. Vertical arrows corresponds to the cases of isentropic nitrogen releases from high pressure to atmospheric pressure of 1 bar. The area under thick black bell-shaped line corresponds to the two-phase (gas-liquid) state of nitrogen.

3.0 EXPERIMENTAL DETAILS

A small-scale DISCHA facility was designed and fabricated to simulate the transient blow-down of a cryogenic gas through a small break. Gaseous and liquid nitrogen were used to substitute gaseous and liquid hydrogen (GH_2 and LH_2). The results will complement the quasi-stationary tests and will provide time-dependent data for verification of the theoretical and numerical models developed in [5]. The schematic of the facility is presented in Fig. 4

The test vessel (blue) is located on a low-friction sledge which is mounted itself to a high-resolution scale (capacity 150 kg, resolution 5 g). The vessel has two discharge openings at different heights to test phase separation during the two-phase blow-down, however only one part is used in a given test. The test is initiated by computer controlled opening of the fast value V5, or V4, respectively. The discharging fluid will create a thrust *F* which can be measured by the force transducer *F*. The scale provides an independent measurement of the mass flow $\dot{m}$ leaving the vessel. Both quantities are connected by the thrust equation:

$$F = \dot{m}V_e + (p_e + p_0)A_e, \qquad (4)$$

where F is the thrust [N]; $\dot{m}$ mass flow rate [kg/s]; V_e is the exit velocity [m/s]; p_e is the exit pressure [Pa]; p_0 is the ambient pressure [Pa]; A_e nozzle area [m^2]. When the exit flow conditions are determined theoretically, Eq. (4) allows to check a consistency of measured and calculated data. The same nozzle geometry is as in the ICESAFE facility [3] is used in these tests.

The piping system shown in Fig. 4 allows to fill the test vessel with gaseous or liquid nitrogen. The vessel is designed for 200 bar, it has a wall thickness of 30 mm and a volume of 2.81 dm^3. For the initial blow-down tests gaseous nitrogen with up to 200 bar was used. For the tests at ambient temperatures the pressure vessel was not thermally insulated.

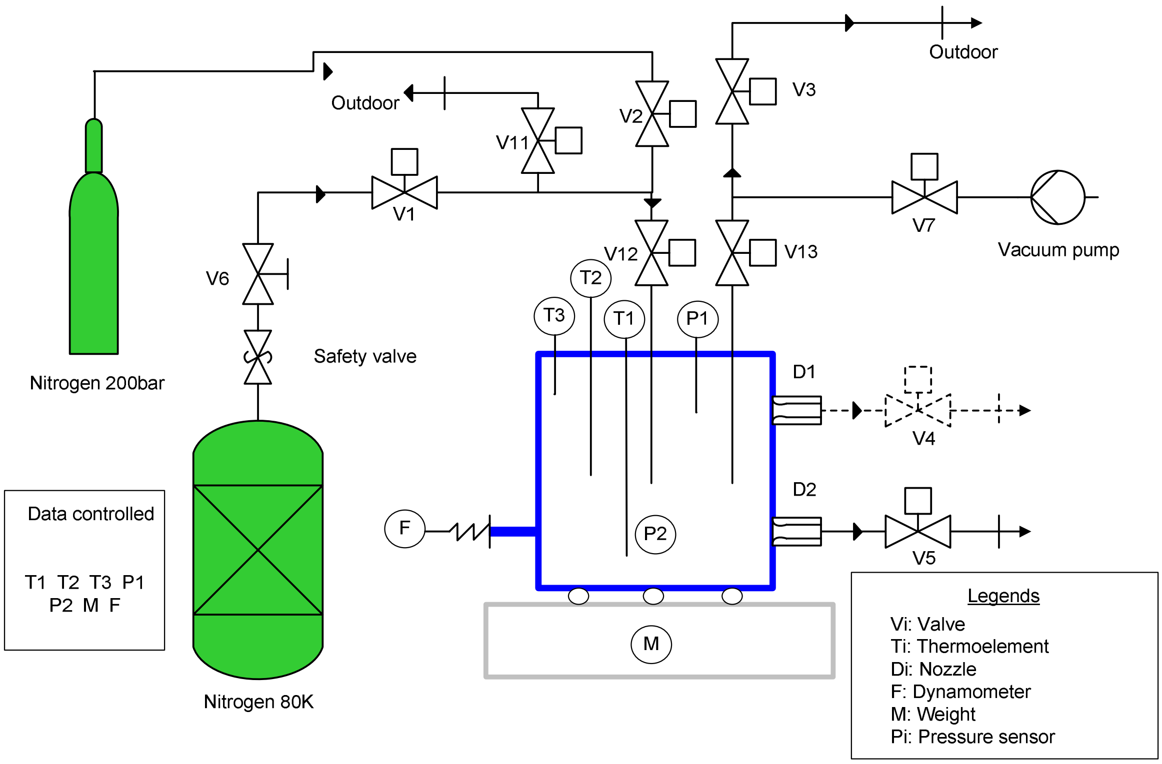

Figure 4. DISCHA facility for transient two-phase blow-down tests with gaseous nitrogen.

The instrumentation is summarized in Fig. 4. It consists of a force transducer (F), the scale (M), three thermocouples inside the vessel at different heights (T1-T3), and two fast pressure transducers inside the vessel at different heights (P1, P2).The test performance, data acquisition, and storage is controlled with an extensive Labview program.

The tests with nitrogen gas were performed with orifice sizes of 0.5, 1, 2 and 4 mm, initial pressures between 30 and 200 bar, initial temperatures between 290 and 306 K, using the upper and lower discharge parts of the vessel D1 and D2 (Fig. 3.75). A calculated by [4] sound speed in nitrogen will be used for scaling measured blow-down pressures. Some experiments were repeated to check the reproducibility of the results. First, the 4 mm – results will be presented as an example for the measurements performed in this project, and then a scaling of measured transient pressures will be performed.

4.0 RESULTS AND DISCUSSION

Fig. 5 (top) displays measured thrust forces for six different initial N_2 pressures. The signals show an exponential decay which is superimposed by an oscillation during the first second. The amplitude of the oscillation increases with increasing initial pressure. In future experiments a stronger support of the force transducer will be tested to remove these oscillations.

The bottom part of Fig. 5 displays the calculated thrust for the 200 bar discharge experiment using Eq. (4). The mass flow $\dot{m}$ is numerically calculated using the relation Eq. (1). Also the exit pressure p_e and the exit velocity V_e are calculated with the adiabatic approach same way as in [5]. The measured thrust can be well reproduced with a discharge coefficient of C_D =0.9. For the type of conical nozzle as used in the present experiments, C_D-values around 0.97 are often quoted. The smaller C_D-value found here may be attributed to the fact, that the calculation assumed an adiabatic discharge of N_2, whereas in the experiment heat transfer from the vessel wall increased temperature and pressure during the blow-down process.

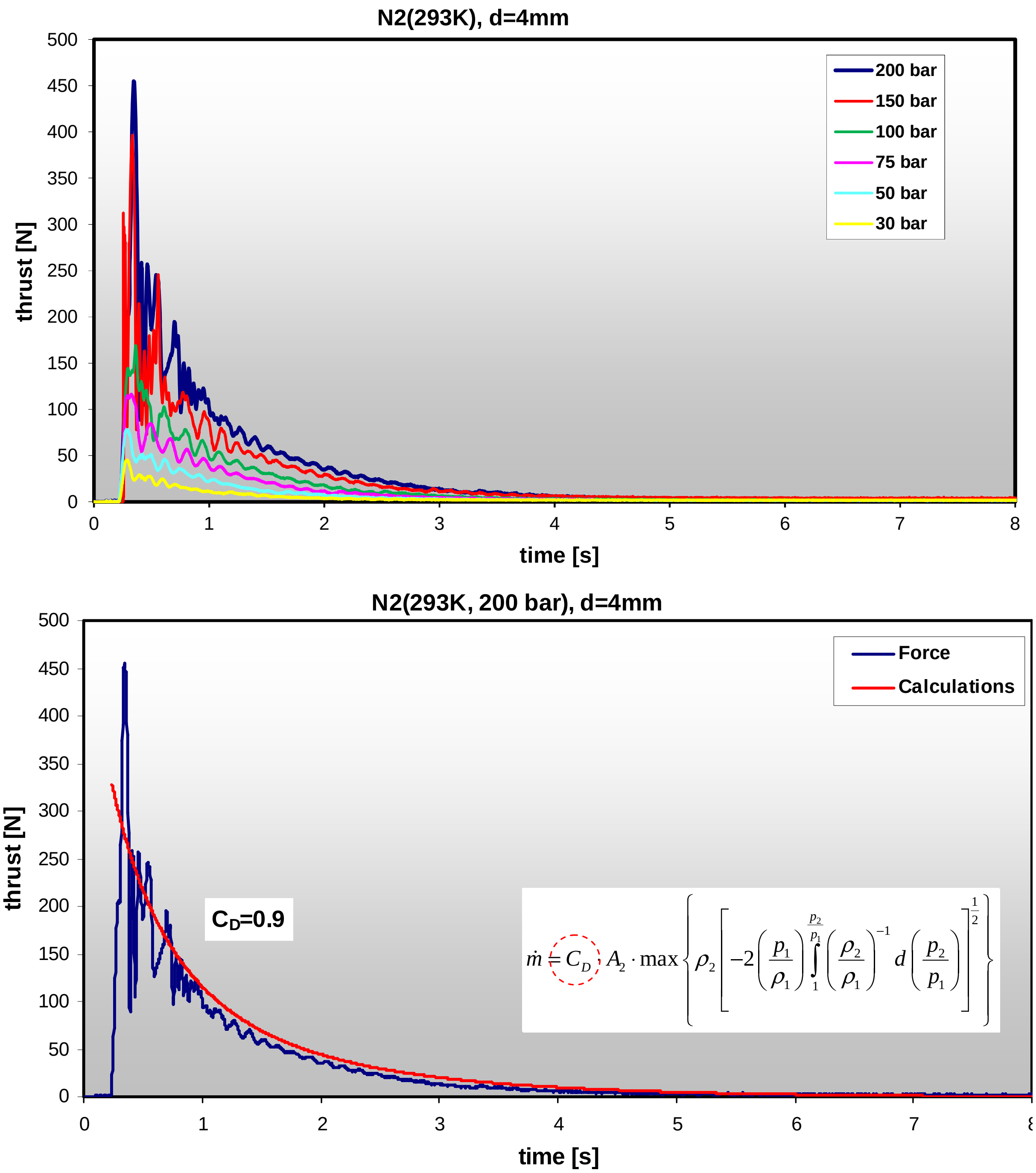

Figure 5. Measured thrust in transient nitrogen gas discharge experiments with different initial pressures (top) and calculated time dependent thrust for the 200 bar experiment (bottom). The orifice size was 4 mm.

Fig. 6 displays the corresponding results for measured and calculated pressures. The measured pressures show the expected smooth decay without any oscillations. This decay has been analyzed in detail assuming ideal gas behaviour and constant values for $\gamma = c_p/c_v$ [6]. Using the nitrogen real gas equation-of-state and the methodology described in [5] for hydrogen it gives very good agreement between experiment and theory, if the same discharge coefficient of 0.9 as in the thrust evaluation is applied.

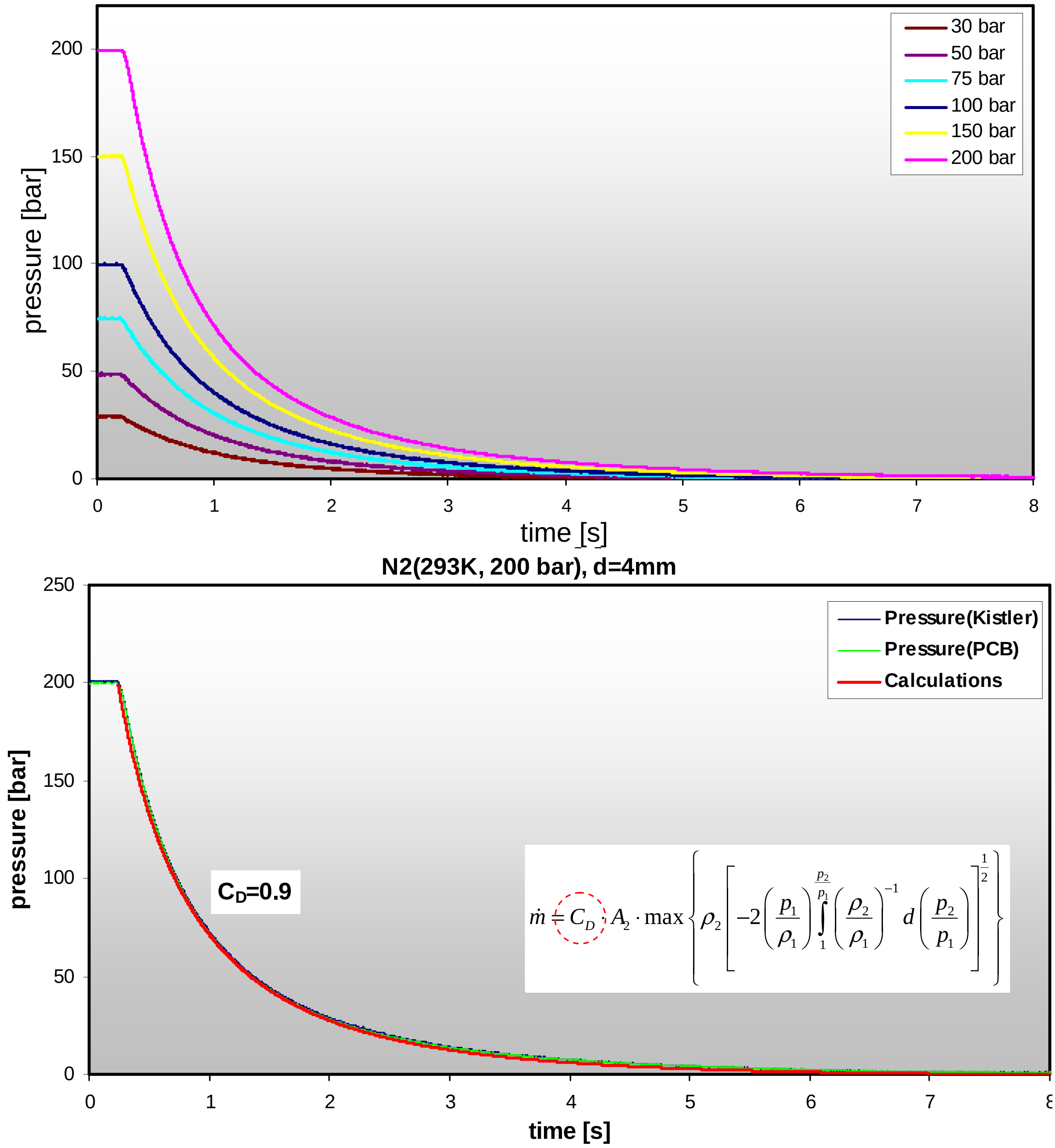

Figure 6. Measured vessel pressure in transient nitrogen gas discharge experiments with different initial pressures (top) and calculated time-dependent pressure for the 200 bar experiment (bottom). The orifice size was 4 mm.

Figure 7 compares measured and calculated transient gas temperatures for a nitrogen discharge experiment starting from 200 bar and 293 K. The nozzle diameter was 4 mm. The calculated adiabatic gas temperature agrees very well with the thermocouple data during the first 2-3 seconds. Thereafter heat transfer from the walls prevents further gas cooling and later even causes warming of the nitrogen gas. Again the discharge coefficient C_D = 0.9 was used in the calculations.

Such a C_D value is consistent with the theoretical results of [6], in which an adiabatic and isothermal discharge of air is compared. The first limiting case assumes no heat transfer from walls, the second limiting case assumes sufficient time for heat transfer to maintain the temperature of the gas in the vessel constant. The predicted mass flow for the isothermal discharge is about 30% smaller than that of the adiabatic case. Since the N_2-experiments described here, present an intermediate case between the two limiting discharge cases, a slight decrease of the discharge coefficient C_D due to wall-gas heat transfer can be expected.

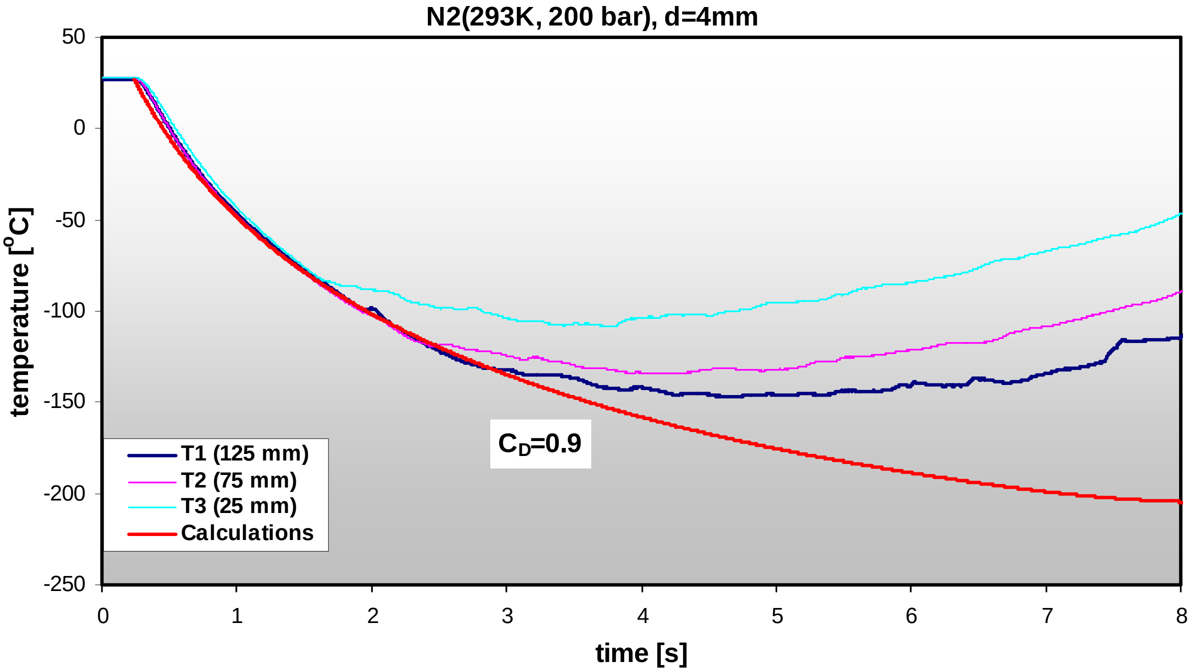

Figure 7. Measured and calculated gas temperatures in a N_2 discharge experiment starting from 200 bar and 293 K. The experimental data indicate heat transfer from vessel walls and thermal stratification. Nozzle diameter 4 mm.

The thermocouple measurements indicate a thermal stratification in the vessel during the blow-down process. The coldest temperatures are recorded at the bottom (thermocouple T1, 125 mm below the vessel cover). Thermocouple T1 is also the closest one to the discharge nozzle D2 used in this test (see Fig. 4).

Heat transfer during the blow-down process was also analyzed by calculating the time-dependent N_2 gas entropy S (p, T) from measured pressures and gas temperatures during the discharge, using the NIST real gas equation-of-state [4]. Figure 8 displays calculated gas entropies for a 5- bar, 50-bar, 100-bar, and 200-bar discharge experiment, using the 4 mm nozzle and the data of thermocouple T1. The initial gas temperature in these tests was near 300 K. The gas entropies are initially constant, following the isentropic path very closely, but later heat transfer causes temperature and entropy increases, preventing the formation of two-phase states, which would occur an ideally isentropic expansion from 200 bar. In the present set-up heat transfer becomes significant after depressurization to about 10% of the initial pressure, except in the 5-bar test which was nearly adiabatic due to the small discharge time and smaller temperature gradient.

The influence of the discharge time becomes very obvious if the experiments from 200 bar initial pressure but with different nozzle diameters (Fig. 9). The blow-down times required from 200 bar to 10 bar were 227 s, 59 s, 14 s, and 3.4 s for the 0.5 mm, 1 mm, 2 mm, and 4 mm nozzles, respectively. The longer the discharge time, the smaller the temperature decrease in the gas due heat transfer from the walls. The response time of the thermocouple, which consisted of bare (unsheathed) 0.1 mm Type K wires, was less than 10 ms.

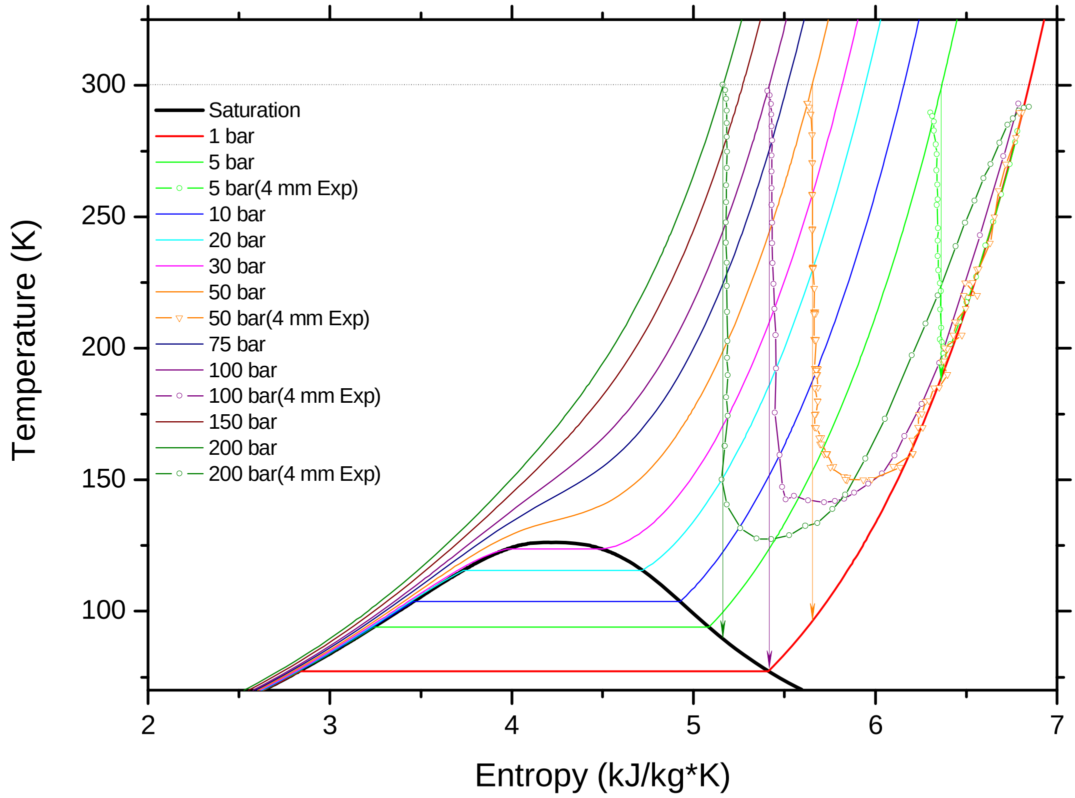

Figure 8. Calculated N2 gas entropies in N2-discharge experiments with 5, 50, 100, and 200 bar initial pressure and 4 mm nozzle diameter. The high pressure tests are influenced by heat transfer from the wall to the gas, which prevents two-phase release for the 100 and 200 bar tests.

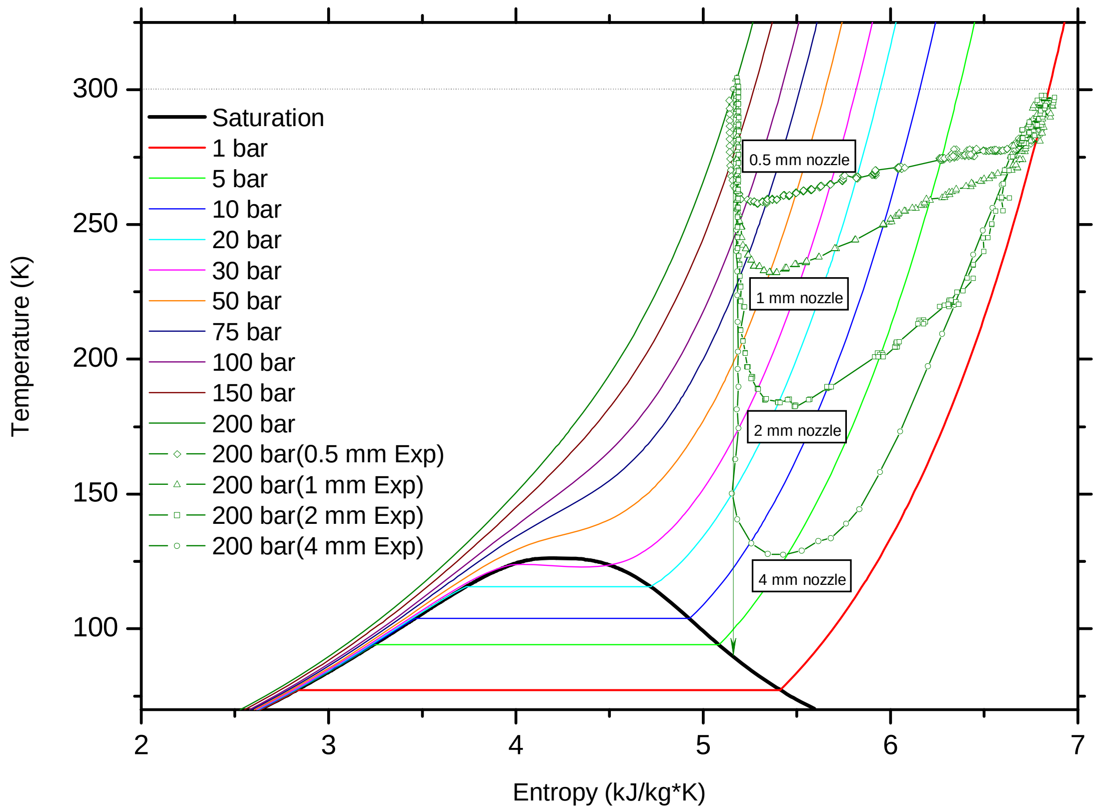

Figure 9. Calculated nitrogen gas entropies in nitrogen discharge experiments from 200 bar initial pressure with different nozzle diameters.

The GN_2 experiments have provided important information on how heat transfer influences the blow-down of a high-pressure gas system. It shows that heat transfer from ambient atmosphere may prevent two phase flow for relative small leaks. This means that it needs larger nozzle diameter, higher pressure, lower initial temperature and better thermal isolation to really reach the two-phase state.

5.0 SCALING OF TRANSIENT DISCHARGE PRESSURES

The transfer of experimental data from any small scale facility to practical applications on larger scale requires a dimensional analysis of the governing processes. For the discharge of ideal gases from higher pressure vessels non-dimensional analytical solutions have been presented for the time-dependent pressure in the reservoir [6]:

$$p^{+} = \left[1 + \left(\frac{\gamma - 1}{2} \right) \left(\frac{\gamma + 1}{2} \right)^{\frac{-(\gamma+1)}{2(\gamma-1)}} \cdot t^{+} \right]^{\frac{-2\gamma}{\gamma-1}}, \quad (5)$$

where t is the time; p is the pressure; dimensionless pressure, $p^{+} = p(t)/p_0$; p_0 is the initial bulk pressure; adiabatic exponent, $\gamma = c_p/c_v$; dimensionless time, $t^{+} = t/t_{char}$; characteristic release time, $t_{char} = V/(A \cdot c_0)$; V is the vessel volume; A is the nozzle area; c_0 is the initial sound speed of gas in vessel. Measured experimental pressures $p(t)$ are displayed in Fig. 10 in terms of p^{+} vs. t^{+} for the different nozzle diameters of 2.0, and 4.0 mm. Scaling with p^{+} and t^{+} results in very good agreement of the tests with different initial pressures for a given nozzle. There is a small systematic spreading of the curves for $p^{+} < 0.1$, which is probably due to gas-wall heat transfer. The measures pressures $p(t)$ were evaluated from the initial pressure p_0 down to p_{end} = 3 bar to remain in the choked flow regime.

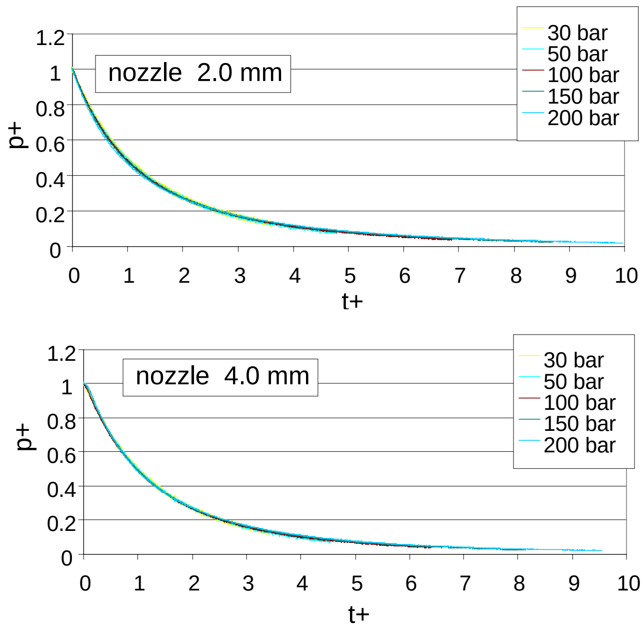

Figure 10. Scaling of measured transient pressures for different initial pressures (30-200 bar) and nozzle diameters (2.0 and 4.0 mm) using the upper discharge port D1.

Note that the used characteristic time includes the sound speed of the gas c_0 in its initial state p_0/T_0, which varies significantly with the initial pressure. Although Eq. (5) was originally derived for ideal gases with constant γ and constant sound speed during the blow-down. It also allows a very good scaling of the present non-ideal high-pressure discharge experiments with nitrogen.

Figure 11 compares the scaled pressure histories for the upper discharge port and for the five different nozzle diameters. For each given nozzle diameter only the largest initial pressure is plotted (30 and 200 bar). A good overall correlation is observed. The differences are due to the above discussed heat transfer effects and the discharge time. The slowest experiments (0.5 and 1 mm nozzles) show the highest values for $p^+(t^+)$. For instance the scaled time t^+ to reach a scaled pressure $p^+ = 0.2$ is about $t^+ = 3.5$ for the 0.5 mm nozzle. This number shifts to smaller values for increasing nozzle diameter and decreasing discharge time, reaching about $t^+ = 2.5$ for the 4 mm nozzle.

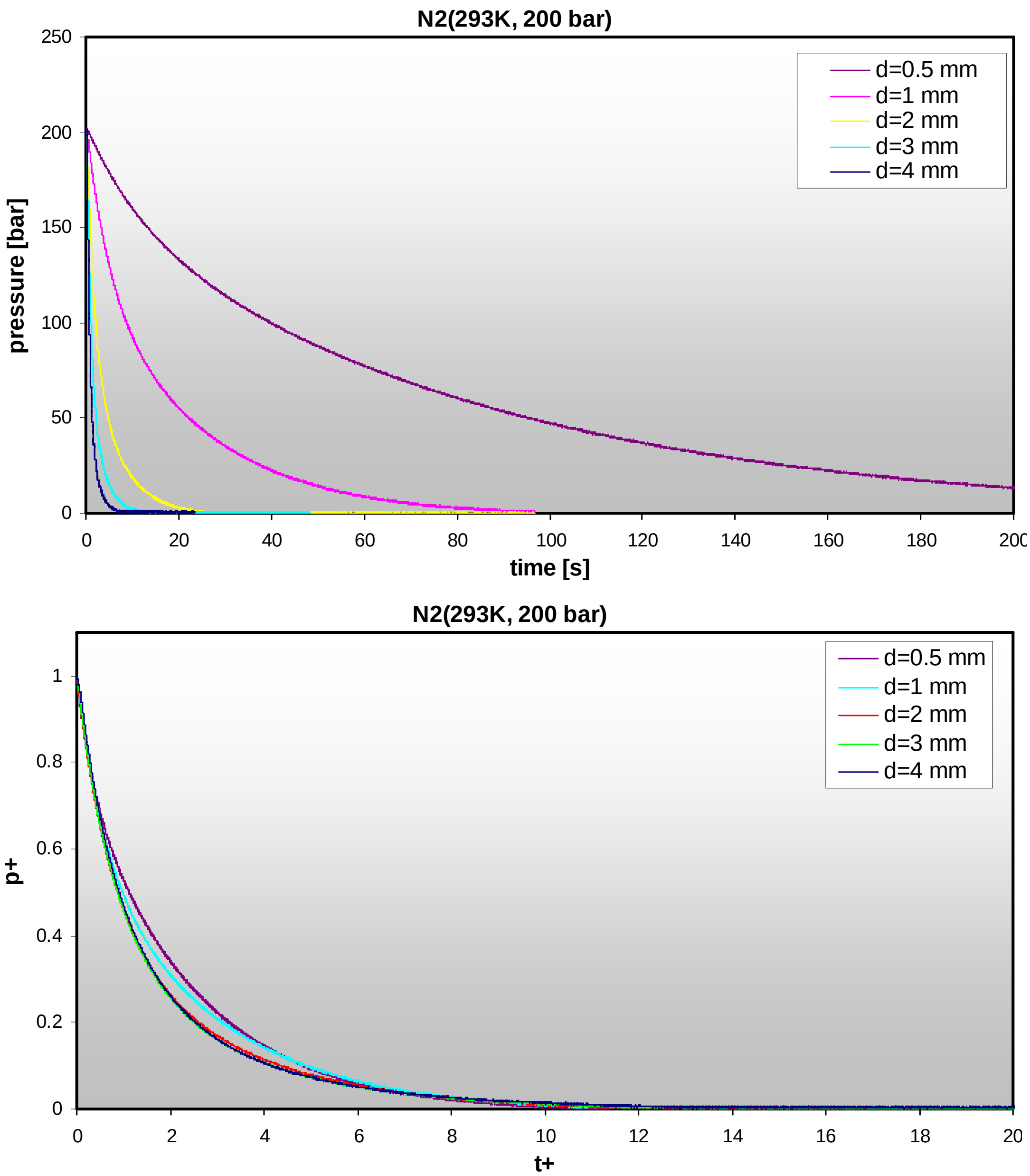

Figure 11. Scaling of measured transient pressures for different nozzle diameters and initial pressure of 200 bar using the upper discharge port D1.

The comparison of experimental results and calculations by Eq. (5) shows that the experimental results with heat transfer tend to converge against the theoretical solution without heat transfer with decreasing discharge time.

The above discussed discharge pressures were all measured using the upper discharge nozzle D1 in Fig. 4. The same test series was performed with the lower discharge nozzle D2 giving identical p(t) within the pressure transducer precision (0.5%). As follows from current experiments, in case of single-phase discharge of nitrogen from the test vessel no effect of the release location on the discharge pressure was observed, and the scaling discussed above leads to practical identical results.

6.0 SUMMARY AND CONCLUSIONS

A small-scale facility for transient discharge of cryogenic nitrogen was designed, constructed and tested with gaseous nitrogen. Different orifice sizes (0.5, 1, 2, 3, 4 mm) and initial N_2 pressures (30 – 200 bar) were investigated.

The measured time-dependent data for vessel discharge pressure, thrust, discharge mass flow, and gas temperatures could be well reproduced using the NIST database for the real gas equation-of-state of nitrogen [4]. This verification for nitrogen also assures the EOS for hydrogen, which is based on the same methodology [5].

Another important finding is, that the newly developed critical discharge analysis method for a pure substance [5] predicts correctly the transient blow-down of a high-pressure gas system. Because a number of physical model assumptions and numerical approximations are involved in the methodology, correctness and precision for transient simulations had to be demonstrated. The dynamic nitrogen tests have provided an additional verification which is complementary to the quasi-static DISCHA experiments.

The measured pressure histories could be scaled very well using initial pressure and sound speed of the gas, vessel volume and nozzle area as characteristic quantities.

New results about heat transfer effects in blow-down of gaseous high-pressure systems have been obtained. For relatively small nozzle diameter, lower initial pressure and absence of thermal isolation the heat from surrounding may completely eliminate the two-phase scenario of high pressure release.

The facility is ready for extension of the experiments to liquid nitrogen and investigation of cryogenic two-phase discharge in a future project phase.

ACKNOWLEDGEMENTS

This work was done within the Icefuel project (www.icefuel.de) funded by the German Federal Ministry of Education and Research.

REFERENCES

1. A. Dylla, H2-transport durch Kabel. Energieversorgung "HZwei –Das Magazin für Wasserstoff und Brennstoffzellen", (April 2007) 7. Jahrgang
2. A. Veser, M. Kuznetsov, G. Fast, A. Friedrich, N. Kotchourko, G. Stern et al. The structure and flame propagation regimes in turbulent hydrogen jets, *International Journal of Hydrogen Energy*, **36**, 2011, pp. 2351–2359
3. A. Friedrich, W. Breitung, G. Stern, A. Veser, M. Kuznetsov, et. al, Ignition and heat radiation of cryogenic hydrogen jets, *International Journal of Hydrogen Energy*, **37**, 2012, pp. 17589-17598.
4. P.J. Linstrom and W.G. Mallard, Eds., NIST Chemistry WebBook, National Institute of Standards and Technology, Gaithersburg MD, 20899, http://webbook.nist.gov, (retrieved May 23, 2013)
5. J. Xiao, J.R. Travis, W. Breitung, Hydrogen release from a high pressure gaseous hydrogen reservoir in case of a small leak, *International Journal of Hydrogen Energy*, **36**, 2011, pp. 2545-2554
6. J. Dutton, R. Coverdill, Experiments to study the gaseous discharge and filling of vessels, *Int. J. Eng. Ed.*, **13** , 1997, pp. 123–134.