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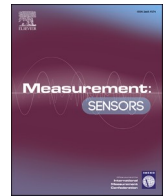
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## Thermodynamic properties of hydrogen containing systems and calculation of gas critical flow factor

### ARTICLE INFO

#### Keywords:

Critical flow factor ( $C^*$ )  
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### ABSTRACT

Sonic nozzles are emerging as crucial reference devices in the calibration of flow meters designed for hydrogen service. In this research study we aim to investigate existing equations governing the critical flow factor ( $C^*$ ) for hydrogen and assess the achievable uncertainty in determining this vital property. An examination of the literature has been undertaken to target experimental measurements related to hydrogen mixtures, setting the stage for a comprehensive gap analysis.

We introduced  $C^*$  values and validated our calculations with two calibration gases: nitrogen, and methane and their standardised  $C^*$  values. Then, the verified methodology has been utilised for generating  $C^*$  values for hydrogen.

This study concluded that the integration of precise experimental data and the utilisation of representative equations and optimised thermodynamic models is essential for enhancing the accuracy of  $C^*$  calculations, particularly in the context of the expanding role of sonic nozzles in hydrogen flow meter calibration.

### 1. Introduction

Many governments worldwide are investigating the potential to reduce carbon dioxide ( $\text{CO}_2$ ) emissions with hydrogen as an energy vector and as an energy buffer for intermittent renewable energy. In each case, there is a need for accurate flow measurement for billing and taxation purposes. This is problematic because there are very few existing traceable, independent flow calibration laboratories for hydrogen.

A technology which is widely used in industry and by national measurement institutes for high accuracy gas flow measurement is the sonic nozzle. Sonic nozzles have many advantages including the fact that they can achieve measurement uncertainties which are slightly higher than those of primary flow standards. They consist of a single element with no moving parts and when operated with care, they demonstrate negligible drift even over extended periods.

An additional advantage to sonic nozzles is that the discharge coefficient is consistent even when different gases are used i.e., a sonic nozzle calibrated with air will perform consistently for methane, provided that accurate fluid property calculations are applied. This potentially allows for sonic nozzles to be calibrated in air and subsequently used for high accuracy flow measurement of hydrogen with only a slight increase in measurement uncertainty. The magnitude of this increase depends on the uncertainty in the calculated critical flow factor ( $C^*$ ) for hydrogen.

Using the international standard ISO 9300 [1], the  $C^*$  for several gases (nitrogen, argon, air, methane, oxygen, and carbon dioxide) can be quickly calculated to an uncertainty of  $\pm 0.1\%$  at 95% confidence but a methodology for calculating the  $C^*$  for hydrogen is not included in the standard. Based on the relatively large uncertainties for other hydrogen thermophysical properties such as density, speed of sound (SoS), and

heat capacities, the uncertainty in the calculation of  $C^*$  for hydrogen could be larger than that of the gases currently included in ISO 9300.

A review was performed based on existing literature available for thermodynamic properties of pure hydrogen and hydrogen mixtures, to highlight areas that existing research covers, and where there are gaps in experimental data.

Owing to the intricate relationships between mass flow rate through a critical nozzle and hydrogen gas thermodynamic properties, a few studies have demonstrated the hydrogen gas flow discharge coefficient. Johnson [2] depicted that the mass flow rate of a real gas, assuming one-dimensional, isentropic flow, through a critical flow nozzle can be expressed in terms of  $C^*$ . Morioka et al. [3] have built a critical nozzle flow meter for high-pressure hydrogen gas flow metering, and its characteristics were experimentally tested with hydrogen gas pressure up to 700 bar. Stewart et al. [4] provided more accurate  $C^*$  values for air, argon, nitrogen, and methane in the low temperature, high pressure range compared to the values reported by Johnson [2] and Schmidt et al. [5].

However, most previous research works struggled with a lack of sufficiently detailed knowledge and understanding of the effect of the real gas state equation. In addition, other researchers formulated some equations to determine the mass flow rate of hydrogen gas through a critical nozzle, but with some lack of uncertainty quantifications on those models. Therefore, a highly accurate equation of state (EoS) for calculation of gas thermodynamic properties needs to be introduced.

In this study, accurate EoS for hydrogen and precise equations for gas flow through a critical nozzle will be presented. Then, the results will be compared with those generated in previous research studies.

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## 2. Methodology

### 2.1. Determination of gas critical flow factor

The ideal critical flow rate for one-dimensional isentropic ideal gas flow,  $q_{mi}$  (mass flow rate), is calculated as follows:

$$q_{mi} = \frac{A_t C_i^* p_0}{\sqrt{R_m T_0}} \quad (1)$$

where  $C_i^*$  is ideal gas critical flow factor and calculated by  $\sqrt{\gamma(2/\gamma + 1)^{\gamma+1/\gamma-1}}$ . This value for hydrogen is equal to 0.68747.

The ideal discharge coefficient ( $C_d^i$ ) is also calculated using the following equation:

$$C_d^i = \frac{q_m \sqrt{R_m T_0}}{A_t C_i^* p_0} \quad (2)$$

The real discharge coefficient,  $C_d^r$ , can be calculated by replacing the ideal gas critical flow factor,  $C_i^*$ , with the real gas critical flow factor,  $C_r^*$ , in Eq. (2) as follows:

$$C_d^r = \left( \frac{C_i^*}{C_r^*} \right) C_d^i = \frac{q_m \sqrt{R_m T_0}}{A_t C_r^* p_0} \quad (3)$$

Therefore, the ideal discharge coefficient ( $C_d^i$ ) is divided into:  $C_d^r$  that accounts for viscous, and  $\frac{C_r^*}{C_i^*}$  that just reflects the impact of real gas. For calculation of  $\frac{C_r^*}{C_i^*}$ , the real gas critical flow factor,  $C_r^*$ , can be determined utilising the following equation:

$$C_r^* = \rho_t V_t \frac{\sqrt{R_m T_0}}{P_0} \quad (4)$$

The enthalpy alteration in a flowing gas is respective to the flow velocity,  $V$ , through  $dh = VdV$ . The following expression is determined by integration of (4) between the upstream stagnation and throat conditions:

$$h_0 - h_t = \frac{V_t^2 - V_0^2}{2} \quad (5)$$

And as the velocity at the throat is equal to the local speed of sound (SoS), the enthalpy and entropy conditions at the throat are:

$$h_t = h_0 - \frac{w_t^2 - V_0^2}{2} \quad (6)$$

$$s_t = s_0 \quad (7)$$

Where  $h_t$  is the enthalpy at the throat condition,  $h_0$  is the enthalpy at the upstream stagnation condition,  $w_t$  is the velocity at the throat condition,  $s_t$  is the entropy at the throat condition,  $s_0$  is the entropy at the upstream stagnation condition.

There is also a correlation for hydrogen  $C^*$  calculations generated by Corpron [6] through fitting a polynomial to available data [2] and it has been mathematically defined as:

$$C^* = A_0 + A_1 \text{Log}\left(\frac{1.8T}{100}\right) + A_2 P + A_3 \left[ \text{Log}\left(\frac{1.8T}{100}\right) \right]^2 + A_4 P \text{Log}\left(\frac{1.8T}{100}\right) + A_5 \left[ \text{Log}\left(\frac{1.8T}{100}\right) \right]^3 + A_6 P \left[ \text{Log}\left(\frac{1.8T}{100}\right) \right]^2 \quad (8)$$

Where  $T$  (K) and  $P$  (bar) are the absolute temperature and pressure, respectively, and  $A_0 = 0.79741185$ ,  $A_1 = -0.33912011$ ,  $A_2 = 0.00029854078$ ,  $A_3 = 0.33862248$ ,  $A_4 = -0.0010015041$ ,  $A_5 = -0.11242827$ ,  $A_6 = 0.00067411915$ .

Within the pressure range of 0 bara to 101 bara, this equation fits the data with accuracy of  $\pm 0.0123\%$ – $95\%$  confidence for the temperature range of 244 K–333 K. For the temperature range of 278 K–333 K, the accuracy of the fit is  $\pm 0.0103\%$ .

### 2.2. Solutions of the equations

The solving of an EoS is an iterative process of solving the enthalpy and entropy balances for the throat conditions. The EoS is utilised to determine the corresponding enthalpy, entropy, and density related to a particular upstream stagnation temperature and pressure. As the EoS is a function of temperature and density, it is more efficient to work with these variables.

The process starts with a primary guess of the throat temperature ( $T_t^{(n)}$ ) and density ( $\rho_t^{(n)}$ ),  $n$  as newest estimated value. Since both  $h$  and  $s$  can be functions of temperature and density, we have on differentiation:

$$\Delta h = \left( \frac{\partial h}{\partial T} \right)_\rho \Delta T + \left( \frac{\partial h}{\partial \rho} \right)_T \Delta \rho \quad (9)$$

$$\Delta s = \left( \frac{\partial s}{\partial T} \right)_\rho \Delta T + \left( \frac{\partial s}{\partial \rho} \right)_T \Delta \rho \quad (10)$$

Then, we can replace the four partial derivatives in terms of thermodynamic properties which can be calculated from the EoS that results in:

$$\Delta h = v^2 \kappa (1 - T\alpha) \Delta \rho + (c_v + v\alpha\kappa) \Delta T \quad (11)$$

$$\Delta s = -v^2 \alpha \kappa \Delta \rho + \left( \frac{c_v}{T} \right) \Delta T \quad (12)$$

Where  $v$  is specific volume,  $\kappa$  stands for isothermal bulk modulus,  $\alpha$  is Helmholtz free energy,  $c_v$  is isochoric specific heat capacity,  $\Delta h$  and  $\Delta s$  are for flow in a critical flow nozzle based on equations (6) and (7) are:

$$\Delta h = h_0 - \left( h_t + \frac{w_t^2 - V_0^2}{2} \right) \quad (13)$$

$$\Delta s = s_0 - s_t \quad (14)$$

Thus, we attain the following equations through integrating equations (11)–(14):

$$h_0 - \left( h_t + \frac{w_t^2 - V_0^2}{2} \right) = v^2 \kappa (1 - T\alpha) \Delta \rho + (c_v + v\alpha\kappa) \Delta T \quad (15)$$

$$s_0 - s_t = -v^2 \alpha \kappa \Delta \rho + \left( \frac{c_v}{T} \right) \Delta T \quad (16)$$

So, the solutions for  $\Delta T$  and  $\Delta \rho$  are as follows:

$$\Delta T = T \frac{(s_0 - s_t)(1 - T_t \alpha_t) + \alpha_t \left[ h_0 - \left( h_t + \frac{w_t^2 - v_0^2}{2} \right) \right]}{c_v + T_t v_t \alpha_t^2 \kappa_t} \quad (17)$$

$$\Delta \rho = \frac{\left[ h_0 - \left( h_t + \frac{w_t^2 - v_0^2}{2} \right) \right] c_{v_t} - T_t (s_0 - s_t) (c_{v_t} + v_t \alpha_t \kappa_t)}{v_t^2 \kappa_t (c_{v_t} + T_t v_t \alpha_t^2 \kappa_t)} \quad (18)$$

All thermodynamic quantities at the nozzle throat including  $h_t$ ,  $s_t$ ,  $w_t$ ,  $c_{v_t}$ ,  $v_t$ ,  $\alpha_t$ , and  $\kappa_t$  are calculated at the newest estimate of the throat density and temperature,  $\rho_t^{(n)}$  and  $T_t^{(n)}$ . Then, the determined values  $\Delta T^{(n)}$  and  $\Delta \rho^{(n)}$  are utilised to determine the improved estimates for the throat density and temperature as follows:

$$\rho^{(n+1)} = \rho^{(n)} + \varphi \Delta \rho^{(n)} \quad (19)$$

$$T^{(n+1)} = T^{(n)} + \varphi \Delta T^{(n)} \quad (20)$$

Where  $\varphi$  is a multiplier with a value close to unity which is employed to accelerate convergence at high pressures. It has been realised that for  $P > 200$  bar, the convergence rate can be doubled.

### 2.3. Equation of state (EoS)

In the iteration for  $C^*$  the required properties are determined from the most precise thermodynamic models available for each fluid. These models have been developed to represent the best data available for the fluid within that data's experimental uncertainty.

Here in this study, the highly accurate GERG-2008 EoS [7,8] was employed to estimate the thermophysical properties of normal hydrogen. The thermodynamic properties of the fluids at certain temperatures ( $T$ ) are based on a multi fluid approximation using dimensionless Helmholtz energy obtained from:

$$\alpha(\delta, \tau, \mathbf{x}) = \alpha^o(\rho, T, \mathbf{x}) + \alpha^r(\delta, \tau, \mathbf{x}) \quad (21)$$

Where  $\mathbf{x}$  is the molar composition vector,  $\tau = T/T_r$  is inverse reduced temperatures, and  $\delta = \rho/\rho_r$  is reduced density both of which are dependent on molar composition vector. The ideal-gas contribution ( $\alpha^o$ ) is related to number of mixture components ( $N$ ), the mole fraction of each component  $i$  ( $x_i$ ), and the dimensionless Helmholtz energy of component  $i$  in the ideal-gas phase ( $\alpha_{oi}^o$ ) by:

$$\alpha^o(\rho, T, \mathbf{x}) = \sum_{i=1}^N x_i [\alpha_{oi}^o(\rho, T) + \ln x_i] \quad (22)$$

The Residual part of the dimensionless Helmholtz energy ( $\alpha^r$ ) consists of two parts; the linear summation of residual part of the reduced Helmholtz free energy of each component  $i$  ( $\alpha_{oi}^r$ ) and so-called departure function ( $\Delta \alpha^r$ ) which is also function of the fluid composition, the inverse reduced mixture temperature, and reduced mixture density.

The Residual part of the dimensionless Helmholtz can be determined as follows:

$$\alpha^r(\delta, \tau, \mathbf{x}) = \sum_{i=1}^N x_i \alpha_{oi}^r(\delta, \tau) + \Delta \alpha^r(\delta, \tau, \mathbf{x}) \quad (23)$$

The advantage of utilising Helmholtz energy in the given form is that all the other thermophysical properties can be derived analytically from terms  $\alpha^o$  and  $\alpha^r$  and their derivatives. One example is isobaric heat capacity which is mathematically expressed as:

$$c_p(\delta, \tau, \mathbf{x}) = R \left[ -\tau^2 (\alpha_{\tau\tau}^o + \alpha_{\tau\tau}^r) + \frac{(1 + \delta \alpha_{\delta\delta}^r + \delta \tau \alpha_{\delta\tau}^r)^2}{1 + 2 \delta \alpha_{\delta\delta}^r + \delta^2 \alpha_{\delta\delta}^r} \right] \quad (24)$$

Where  $R$  is the universal gas constant. Both  $\alpha^o$  and  $\alpha^r$  show order of their derivatives with respect to  $\tau$  and  $\delta$ . For examples  $\alpha_{\tau\tau}^r$  depicts second order derivatives of  $\alpha^r$  with respect to  $\tau$ . Similarly, enthalpy (h), entropy (s), Gibbs free energy (g), pressure (P) can be attained as follows:

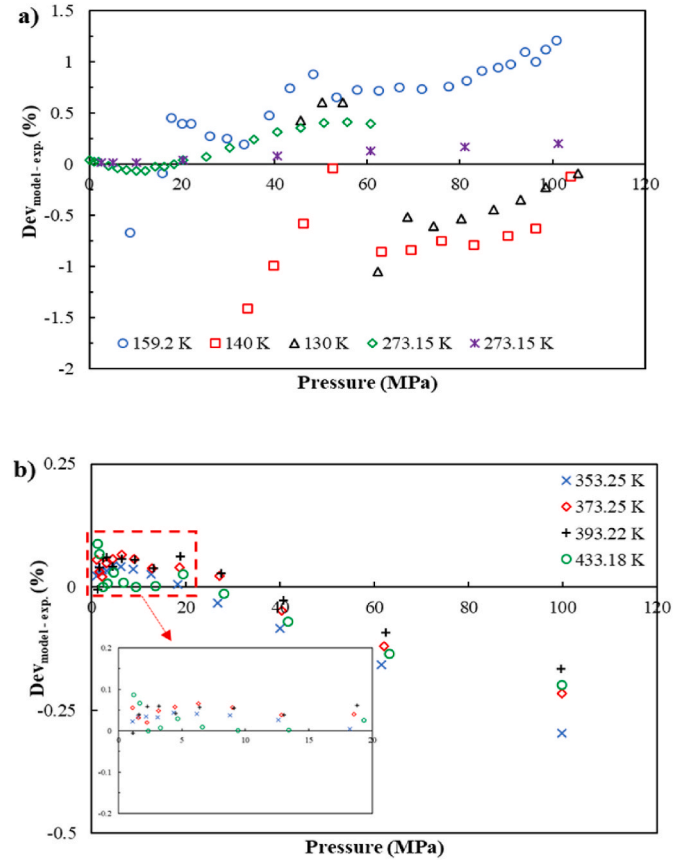


Fig. 1. Deviations between normal hydrogen experimental data on densities [9–13] and determined ones by the EoS used in this study for variety of pressures and temperatures.

$$P(\delta, \tau, \mathbf{x}) = RT\rho [1 + \delta \alpha_{\delta}^r] \quad (25)$$

$$h(\delta, \tau, \mathbf{x}) = RT [1 + \tau (\alpha_{\tau}^o + \alpha_{\tau}^r) + \delta \alpha_{\delta}^r] \quad (26)$$

$$s(\delta, \tau, \mathbf{x}) = R [\tau (\alpha_{\tau}^o + \alpha_{\tau}^r) - \alpha^o - \alpha^r] \quad (27)$$

$$g(\delta, \tau, \mathbf{x}) = RT [1 + \alpha_{\tau}^o + \alpha_{\tau}^r + \delta \alpha_{\delta}^r] \quad (28)$$

Other thermodynamic properties including but not limited to speed of sound and Joule-Thomson coefficient can be described similarly. Kunz. et al. [7] present comprehensive coverage of these derivatives and

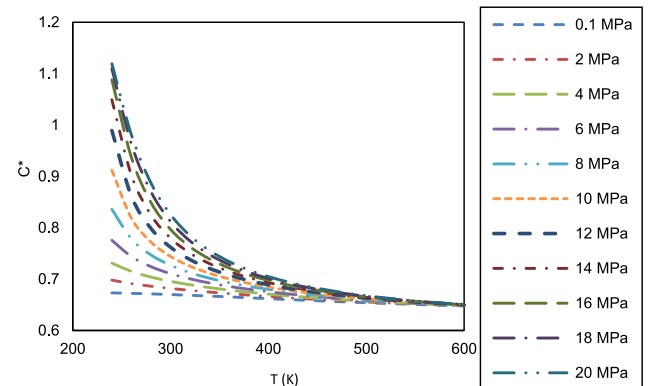


Fig. 2. Critical flow factor for methane.

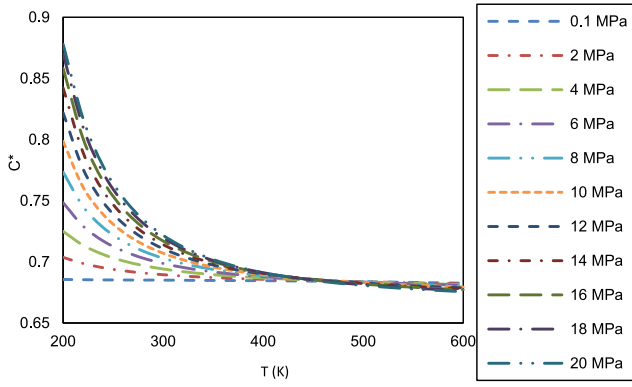


Fig. 3. Critical flow factor for nitrogen.

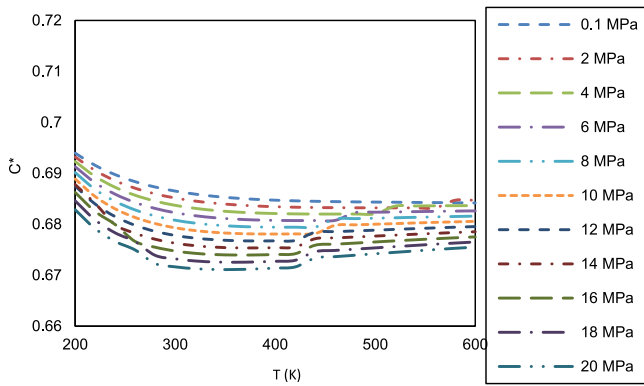


Fig. 4. Critical flow factor for hydrogen.

thermophysical properties.

### 3. Results and discussion

#### 3.1. Comparison of determined normal hydrogen data to some available experimental data

Fig. 1a and b depict deviations of densities plotted versus pressure, separated into temperature increases. As can be seen, the employed EoS displays very good agreement with some available experimental data over a wide range of pressures, i.e.,  $\pm 1.5\%$  for very low temperature conditions and  $\pm 0.5\%$  for higher temperatures. For temperatures higher than 273.15 K, the deviations (Dev.) between modelling and experimental data are mainly within the range of 0.001 %–0.1 % for pressures up to 20 MPa.

#### 3.2. Critical flow factor

The critical flow factors for methane, nitrogen, and hydrogen are given in Figs. 2–4. As can be seen, the values reduce with temperature increments for all pressures.

The critical flow factor for methane and nitrogen depicts an increment with pressure at low temperatures. As the temperature increments, the rate of  $C^*$  increase with pressure increment is decreased. At even higher temperatures the critical flow factor is an inverse function of pressure. This trend was also observed by Stewart et al. [4], who presented the critical flow factors for four calibrations gases: air, argon, nitrogen, and methane, over a wider range of temperatures and pressures (i.e., 200 K–600 K, and up to 20 MPa).

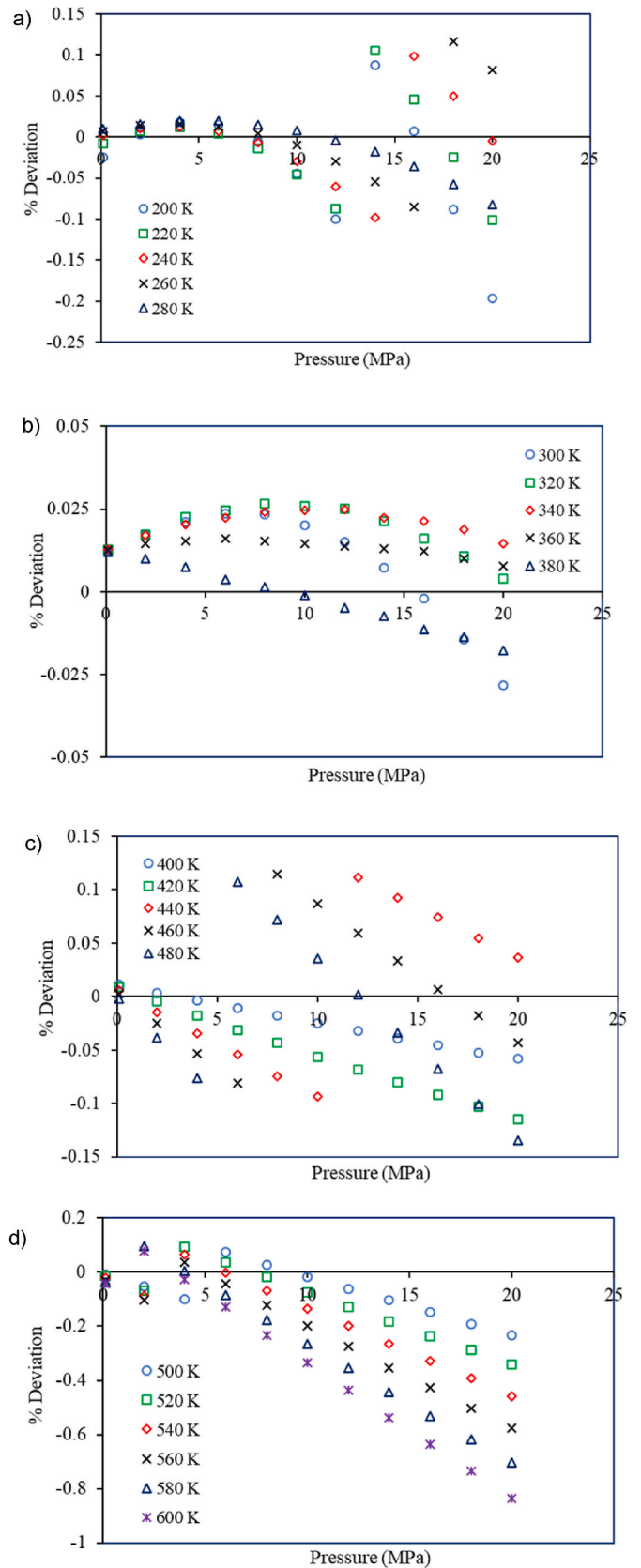


Fig. 5. Comparisons between new  $C^*$  values and Corpron [6] for Hydrogen.

The ISO 9300 standard [4] also gives  $C^*$  values for these four gases in the range from 323.15 K to 373.15 K and up to 10 MPa. The new values for methane and nitrogen are in very good agreement with those of Stewart et al. [4] at low pressures, the differences being less than 0.01 % up to 10 MPa at temperatures of 280 K and above. For nitrogen, the deviations are within the range of 0.001 %–0.01 % at low pressures and abovementioned temperature range. However, as the pressure increments the deviations become larger at temperatures higher than 300 K.

Fig. 5 presents the deviations between new hydrogen critical flow factor values and those from Corpron [6] for wide range of temperatures from 200 K to 600 K and pressures up to 20 MPa.

For temperatures lower than 420 K, the absolute deviations range from 0.01 % to 0.05 % for pressures below 10 MPa. Within the temperature range of 280 K–400 K, the absolute deviation values are mainly in the same acceptable range of 0.01 %–0.05 % for pressures up to 20 MPa. The deviations values increase with both pressure and temperature increments for temperatures increments higher than 360 K.

As can be seen, the critical factor for hydrogen appears to be much less sensitive to temperature and pressure changes than Methane and Nitrogen. The potential reason behind this observation is that the thermophysical properties changes for hydrogen at different pressures and temperatures are less sensitive compared to thermodynamic properties variations of methane and nitrogen at the same pressure and temperature changes. Therefore, the critical flow factor for hydrogen, which is highly dependent on the thermophysical properties of the gases, would be less sensitive to pressure and temperature variations compared to the other two gases.

In the temperature/pressure range previously covered in the literature, the researchers believe that the new values to be more reliable as they are based on more recent and reliable formulations [4,5,14]. Moreover, the greatest differences in  $C^*$  values in comparison to the previous values for nitrogen and methane gases are in the low-temperature and high-pressure region, where the effect of the critical point can be observed. While for Hydrogen gas, the deviations between new and previous  $C^*$  values are greater at higher temperature regions.

The new formulations were developed to represent the thermodynamic properties of fluids in the extended critical regions more precisely. We could see that the lower-pressure conditions lower than 10 MPa represent the most precise values available at present times. However, the deviations are also acceptable for higher pressures up to 20 MPa and temperatures lower than 400 K. If we have access to further thermodynamic experimental datasets in the specific critical regions, then more precise thermodynamic models and EoSs could be developed to attain  $C^*$  values with higher accuracy.

Critical flow nozzles are commonly operated with expected measurement uncertainty of 0.2 %–0.3 % in mass flow rate, at this level, uncertainty in  $C^*$  can be major contributor to the mass flow rate uncertainty. The differences between the new hydrogen  $C^*$  values and previously published values can be noticeable at specific temperatures and pressures.

If a nozzle is calibrated and utilised with the same gas at the same conditions, no error would happen because of utilising inaccurate  $C^*$  values. However, if a nozzle is calibrated with one gas and subsequently employed with another gas then there might be an error in the calculated mass flow rate. This might occur if a nozzle to be employed on hydrogen was calibrated in nitrogen or air. These errors would be integrated to cause errors of up to 0.5 %.

## 4. Conclusions

In this study, many papers for pure hydrogen and hydrogen blends with main impurities have been reviewed to setup this experimental database and provide facility for the potential gap analysis in thermodynamic experimental data of pure hydrogen and hydrogen mixtures. Given the large amount of data gathered from different sources, a gap analysis can be carried out as originally intended. It can be inferred that no specific gap has been identified for density of pure normal hydrogen so far. Generating SoS experimental data for temperature and pressure ranges from 15 K to 350 K and 0.1 MPa–100 MPa would be required in the future. Therefore, in the next stage of this project in future years, an experimental programme will be required to collect the data to fill these gaps.

The new values for the  $C^*$  presented here for nitrogen, methane, and more particularly normal hydrogen are based on the most reliable and optimised EoSs. These equations represent the fluids thermophysical properties within their experimental uncertainties. At lower temperatures and relatively higher pressure ranges these equations are more precise compared to those available in the literature. Thus, it is believed that the  $C^*$  values presented here are the most precise values currently existed. The representative equations and optimised thermodynamic models will give the most precise  $C^*$  values available and accordingly the determination of the theoretical gas mass flow rate through a critical flow nozzle would become easier and more reliable with lower uncertainties.

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