

ESTIMATION OF FINAL HYDROGEN TEMPERATURE FROM REFUELING PARAMETERS

Xiao, J.S.^{1,2}, B nard, P.², and Chahine, R.²

¹ Hubei Key Laboratory of Advanced Technology for Automotive Components and Hubei Collaborative Innovation Center for Automotive Components Technology, Wuhan University of Technology, Hubei 430070, China

² Hydrogen Research Institute, Universit  du Qu bec   Trois-Rivi res, QC G9A 5H7, Canada
E-mails: Jinsheng.Xiao@uqtr.ca, Pierre.Benard@uqtr.ca, Richard.Chahine@uqtr.ca

ABSTRACT

The compressed hydrogen storage is currently widely used in fuel cell vehicles due to its simplicity in tank structure and refueling process. For safety reason, the final gas temperature in the hydrogen tank during vehicle refueling must be controlled under a certain limit, e.g., 85 C. Many experiments have been done for finding the relations between the final gas temperature in the hydrogen tank and different refueling conditions. The analytical solution of the hydrogen temperature in the tank can be obtained from the simplified thermodynamic model of a compressed hydrogen storage tank, and it serves as function formula to fit experimental temperatures. From the analytical solution, the final hydrogen temperature can be expressed as a weighted average form of initial temperature, inflow temperature and ambient temperature based on the rule of mixtures. The weighted factors are related to other refueling parameters, such as initial mass, initial pressure, refueling time, refueling mass rate, average pressure ramp rate (APRR), final mass, final pressure, etc. The function formula coming from the analytical solution of the thermodynamic model is more meaningful physically and more efficient mathematically in fitting experimental temperatures. The simple uniform formula, inspired by the concept of the rule of mixture and its weighted factors obtained from the analytical solution of lumped parameter thermodynamics model, is representatively used to fit the experimental and simulated results in publication. Estimation of final hydrogen temperature from refueling parameters based on the rule of mixtures is simple and practical for controlling the maximum temperature and for ensuring hydrogen safety during fast filling process.

Keywords: hydrogen storage; refueling; fast filling; thermodynamics; temperature; rule of mixtures

NOMENCLATURE

APRR	average pressure ramp rate, MPa/min	\dot{m}_{in}	mass inflow rate for charge, kg/s
a_f	heat transfer coefficient between hydrogen and ambient fluid, W/m ² /K	\dot{m}_{out}	mass outflow rate for discharge, kg/s
A_s	internal surface area of tank, m ²	p_0	initial pressure, MPa
c_p	constant-pressure specific heat, J/kg/K	\dot{p}	APRR, MPa/min
c_v	constant- volume specific heat, J/kg/K	q	specific heat inflow, $q = \dot{Q} / \dot{m}$, J/kg
h	specific enthalpy of hydrogen, J/kg	\dot{Q}	heat inflow rate, $\dot{Q} = a_f A_s (T_f - T)$, W
h_{in}	specific enthalpy of inflow H ₂ , J/kg	t	time variable or fill time, s
h_{out}	specific enthalpy of outflow H ₂ , J/kg	t^*	characteristic time, $t^* = m_0 / \dot{m}$, s
h_∞	specific enthalpy of either inflow or outflow hydrogen, J/kg	T	temperature of hydrogen in tank, K
m	mass of hydrogen mass in tank, kg	T_0	initial temperature in tank, K
m_0	initial hydrogen mass, kg	T_f	temperature of ambient fluid, K
\dot{m}	mass flow rate, $\dot{m} = \dot{m}_{in}$ for charge, $\dot{m} = -\dot{m}_{out}$ for discharge, kg/s	T^*	characteristic temperature, K
		T_∞	inflow or outflow temperature, K
		u	specific internal energy, J/kg

α	dimensionless heat transfer coefficient, $\alpha = a_f A_s / c_v / \dot{m}$	μ	fraction of initial mass over total mass of hydrogen, $\mu = m_0 / m$
γ	ratio of specific heats, $\gamma = c_p / c_v$	τ	dimensionless time, $\tau = t / t^*$

1 INTRODUCTION

Compressed hydrogen storage is currently the preferred means for storing hydrogen in fuel cell vehicles due to its simpler tank and refueling process. For safety reasons, the final gas temperature in the hydrogen tank during refueling is limited to 85 °C. Many experiments have been done for determining the final gas temperature in the hydrogen tank as a function of refueling conditions [1-9]. Numerical simulations [10-16] based on computational fluid dynamics (CFD) have been performed and compared with experiments. Dicken and Mérida [1] proposed a three-parameter formula to fit the relation between the final temperature and the final hydrogen mass in the tank. This formula was used to represent overall CFD simulation results [13]. Thermodynamic properties, such as temperature and pressure evolutions, of hydrogen storage systems are important to evaluate and optimize their performance and to help to choose among different options for the applications [17]. Some researchers also conducted thermodynamic analyses for hydrogen storage system [18-22]. Yang et al carried out an analysis of thermodynamic processes involving hydrogen [18] and a thermodynamic analysis of refueling of a hydrogen tank [19]. Ahluwalia et al analyzed the dynamics of cryogenic hydrogen storage [20] and cryo-adsorption hydrogen storage [21] for automotive applications. Kumar et al developed a lumped parameter model for cryo-adsorber hydrogen storage tank [22]. The authors of the current article have developed a lumped parameter model for charge-discharge cycle of adsorptive hydrogen storage system [23], and have applied it to a cryo-adsorptive hydrogen storage system [24].

Recently, we developed the analytical solutions for the lumped parameter thermodynamic models of compressed and adsorptive hydrogen storage systems [25-26]. The analytical solution of the hydrogen temperature in the tank is used to fit the experimental temperatures. Based on the analytical solution of hydrogen temperature, a two-parameter formula is proposed to replace the three-parameter formula of the reference [1]. Further, a three-parameter formula is proposed to represent three sets of data, which were fitted by triple three-parameter formulae and needed totally nine parameters in the reference [1]. The analytical solution of the hydrogen temperature was successfully applied for the estimation of final hydrogen temperature based on the fitting of experimental temperatures under different final/initial mass ration and different fill times [27].

A general rule of mixtures is a weighted average method used to predict various properties of a composite materials, porous media and multiphase system. It provides a theoretical upper and lower bounds on properties such as the elastic modulus, Poisson's ratio, thermal conductivity and thermal expansion coefficient [28]. The effective heat flux method based on the hypothesis of equivalent temperature difference and the effective temperature difference method based on the hypothesis of equivalent heat flux are proposed for determining the upper and lower bounds of the thermal conductivity of ceramic/metal composite materials [28].

The rule of mixtures may estimate effective/equivalent values of state variables of physics, such as density (as both material prosperity and state variable), stress and temperature. An example for the stress estimation was given in the reference [29], while this study will estimate the final hydrogen temperature in a tank from refueling parameters.

First, we derive the analytical solution of hydrogen temperature from thermodynamic balance equations. Second, the formula of the analytical solution of hydrogen temperature is found being analogous with the rule of mixtures. The thermodynamic basis for estimation procedure of hydrogen temperature is then given. The expression of the fitting results in our recent work [27] will be represented here inspired by the rule of mixtures. Moreover, we will express the final hydrogen temperature as functions of (1) inlet, initial and ambient temperatures, (2) initial pressure and average pressure ramp rate (APRR) and (3) initial pressure, ambient temperature and mass flow rate.

2 THEORY AND METHODS

2.1 General rule of mixtures

For a two-phase system, the upper bound (Voigt model) and lower bound (Reuss model) of a material property P of the mixture can be estimated based on the volume fraction f and simply expressed as

$$P = f_1 P_1 + f_2 P_2 = f_1 P_1 + (1 - f_1) P_2 \quad (1)$$

$$\frac{1}{P} = \frac{f_1}{P_1} + \frac{f_2}{P_2} = \frac{f_1}{P_1} + \frac{1 - f_1}{P_2} \quad (2)$$

These two formulae represent two bounds of the rule of mixtures. The upper bound is mathematically simpler and is more widely used than the lower bound. For density and heat capacity, the upper bound is a good enough estimation. For thermal conductivity, the lower bound may be necessary for some structures of composites or porous media. The effective (moderate) temperature of the mixture (cold and warm hydrogen, or hydrogen and tank wall, or even hydrogen and adsorbent) can be estimated based on the equivalent energy method (energy balance method). The energy, like density and heat capacity, is not sensitive with the structure of the mixture or composites. Therefore, the upper bound (Voigt model) of the rule of mixtures is more suitable to estimate the temperature of the mixture.

2.2 Analytical solution of hydrogen temperature from thermodynamic balance equations

From Appendix A, we have obtained the analytical solution of hydrogen temperature from mass and energy balance equations as [25, 27]

$$\frac{T^* - T}{T^* - T_0} = \left(\frac{1}{1 + \tau} \right)^{1+\alpha} \quad (3)$$

where $\tau = t/t^*$ is dimensionless time, $t^* = m_0/\dot{m}$ is characteristic time, T_0 is initial temperature, and a characteristic temperature is defined as [25, 27]

$$T^* = \frac{\gamma T_\infty + \alpha T_f}{1 + \alpha} \quad (4)$$

The solution of mass balance equation for the constant charging flow rate can be written as

$$\frac{m}{m_0} = 1 + \frac{\dot{m}}{m_0} t = 1 + \frac{t}{m_0/\dot{m}} = 1 + \frac{t}{t^*} = 1 + \tau \quad (5)$$

where m_0 is initial mass in the tank, \dot{m} is charging flow rate. Eq.(3) becomes:

$$\frac{T^* - T}{T^* - T_0} = \left(\frac{m_0}{m} \right)^{1+\alpha} = \mu^{1+\alpha} = \mu' \quad (6)$$

where $\mu = m_0/m$ is the fraction of *initial* mass over *final* mass of hydrogen **in the tank**, and $\mu' = \mu^{1+\alpha}$ is the modified initial mass fraction by considering heat transfer which causes $\mu' < \mu$. Thus Eq.(6) becomes a weighted averaging formula:

$$T = \mu' T_0 + (1 - \mu') T^* \quad (7)$$

The dimensionless heat transfer coefficient α is always positive, so that the modified initial mass fraction μ' monotonically increases with the fraction μ . From Eq.(7), we can see that larger *initial* mass fraction μ (subsequently, larger modified initial mass fraction μ') leads to a larger contribution of T_0 than the characteristic temperature T^* to the final temperature T . As just mentioned, the heat transfer causes $\mu' < \mu$, so it will weaken the contribution from the initial temperature T_0 but will count more contribution from characteristic temperature T^* (combination of inlet temperature T_∞ and ambient temperature T_f). Eq.(7) is similar to Eq.(1) in mathematic form, where $f = \mu'$. Therefore, the analytical solution of hydrogen temperature is analogy with the rule of mixtures.

2.3 Thermodynamic basis for the analogy for hydrogen temperature estimation

We assume negligible heat capacity of the wall of the tank and constant specific heats of hydrogen, as in Section 2.2. Furthermore, we assume the tank is adiabatic and its thermal resistance is negligible. The energy balance between initial status and final status can then be written as

$$mc_v T = m_0 c_v T_0 + (m - m_0) c_p T_\infty \quad (8)$$

Dividing by mc_v , we obtain

$$T = \mu T_0 + (1 - \mu) T^* \quad (9)$$

where $T^* = \gamma T_\infty$. The above equation is the simplified form of Eq.(7) for the adiabatic case ($\alpha = 0$). Therefore, the rule of mixtures for final hydrogen temperature basically represents the energy balance of hydrogen in the tank between initial status and final status. The weighted factor of the rule of mixtures μ is the so-called *initial mass fraction*.

Let us consider another example of thermodynamic systems. We keep the adiabatic assumption for simplification but consider the heat capacity of the tank wall. Now the thermodynamics system includes the hydrogen in the tank and the wall of the tank, so the energy balance between initial status and final status of this system becomes

$$mc_v T + m_w c_w T_w = m_0 c_v T_0 + m_w c_w T_{w0} + (m - m_0) c_p T_\infty \quad (10)$$

If the conductive resistance of the tank wall is neglectable, the wall temperatures equal to the hydrogen temperatures, i.e., $T_w = T$ and $T_{w0} = T_0$. Thus, Eq.(10) gives the final temperature as

$$T = f_{MC} T_0 + (1 - f_{MC}) T^* \quad (11)$$

where $f_{MC} = (m_0 c_v + m_w c_w) / (mc_v + m_w c_w)$ is the fraction of initial heat capacity to final heat capacity of the system, and $T^* = \gamma T_\infty$. The final hydrogen temperature is the weighted average of initial temperature and a characteristic temperature, which relates to the inflow enthalpy of hydrogen. The weighted factor of the rule of mixtures corresponds to the *initial heat capacity fraction* in this context. It reduces to $f_{MC} = m_0 / m = \mu$ if the heat capacity of the tank wall $m_w c_w$ is negligible.

3 RESULTS AND DISCUSSIONS

3.1 Effect of initial and final mass on final hydrogen temperature

Divided by initial temperature, the solution (7) becomes

$$\frac{T}{T_0} = \mu' + (1 - \mu') \frac{T^*}{T_0} \quad \text{or} \quad \frac{T}{T_0} = \frac{T^*}{T_0} - \left(\frac{T^*}{T_0} - 1 \right) \mu' \quad (12)$$

Using the modified initial mass fraction from Eq.(6), Eq.(12) becomes a two-parameter formula:

$$\frac{T}{T_0} = A - (A - 1) \left(\frac{m}{m_0} \right)^{-C} \quad (13)$$

where $A = T^* / T_0$, $C = 1 + \alpha$. The data used for the fitting of Eq.(13) is come from the formula [1]:

$$\frac{T}{T_0} = \left[A + B \left(\frac{m}{m_0} \right)^{1/2} \right]^{-C} \quad (14)$$

This formula requires nine parameters to represent the temperature curves for three fill times, as shown in Table 1. We take these results as reference data. We use the derived two-parameter formula (13) to represent experimental data that the three-parameter formula (14) generated. The fitted parameters and the fitted results obtained by two-parameter formula (13) are shown in Table 2 and Figure 1 respectively. Figure 1 shows good agreement. The derived two-parameter formula (13) has same ability to represent experimental data with the three-parameter formula (14). We note that the

lower bound for non-negative heat transfer coefficient must used for fitting the data at $t=190s$. Without setting the lower bound (zero), we will obtain fitting result $C<1$, which means that the heat transfer coefficient is negative. The function formula based on the idea of the rule of mixtures and resulted from the analytical solution of the thermodynamic model is more meaningful physically and more efficient mathematically in fitting experimental temperatures.

Table 1. Parameters of reference formula [1]

Fill Time (s)	A	B	C
40	-35.17000	36.16000	0.04595
190	-9.50400	10.50000	0.05526
370	-10.51000	11.50000	0.04841

Table 2. Parameters of two-parameter formula

Fill Time (s)	Fitted Parameter	Parameter Value	Standard Error
40	A	1.20418	0.00268
	C	1.63112	0.06587
190	A	1.19469	0.00267
	C	1.00000*	0.02833
370	A	1.17326	0.00240
	C	1.02611	0.02985

Formula: $z=A-(A-1)*y^{-C}$, where y : Final/Initial Mass, z : Final/Initial Temperature
 * Lower bound for non-negative heat transfer coefficient

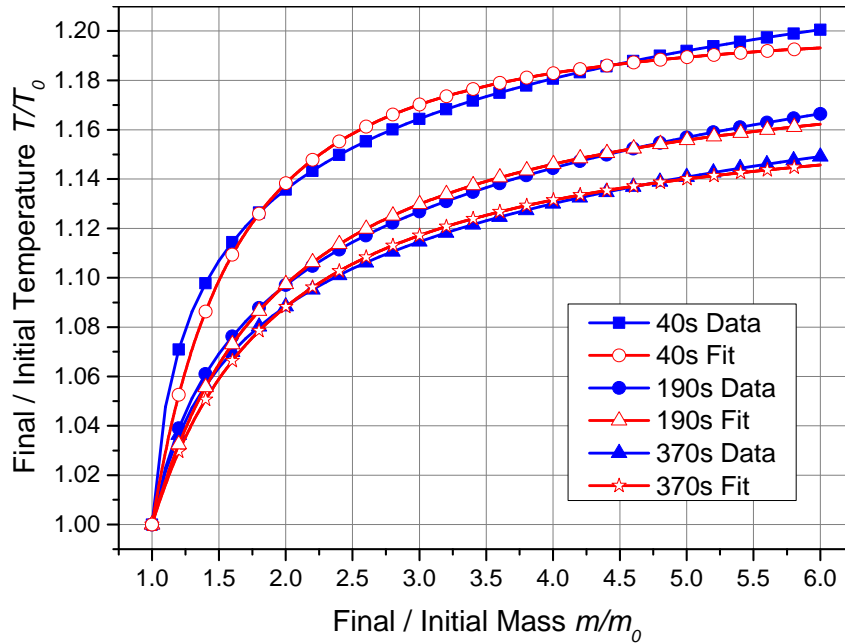


Figure 1. Fitted results of two-parameter formula compared with reference data [1]

3.2 Effect of inlet and initial temperatures

Substituting Eq.(4) into Eq.(7) gives the final temperature as initial, inlet and ambient temperatures:

$$T = \mu' T_0 + \gamma' T_\infty + \alpha' T_f \quad (15)$$

where the modified initial mass fraction, a modified specific heat ratio and a modified dimensionless heat transfer coefficient are denoted as

$$\mu' = \mu^{1+\alpha}, \quad \gamma' = \frac{1-\mu'}{1+\alpha} \gamma, \quad \alpha' = \frac{1-\mu'}{1+\alpha} \alpha \quad (16)$$

The final hydrogen temperature becomes a weighted average of initial, inlet and ambient temperatures: For the experimental data [7], the fitted parameters are shown in Table 3, the fitted results are shown in Figure 2. Considering $T_f = T_0$, we set $\alpha = 0$ to fit a plane and obtain a linear function: $T = 0.59853T_0 + 0.60736T_\infty$. Figure 2 shows that the data points are close to the fitted plane. The factor 0.60736 for the inlet temperature agrees with the number of 0.6 from linear curve fitting [7].

Table 3. Fitted parameters of final temperature function as inlet and initial temperatures

Parameter	Value	Standard Error
mu	0.59853	0.00562
gamma	1.51284	0.00541

Equations: $\mu_1 = \mu^{(1+\alpha)}$; $T = \mu_1 T_0 + (1 - \mu_1) / (1 + \alpha) (\gamma T_\infty + \alpha T_0)$; $\alpha = 0$

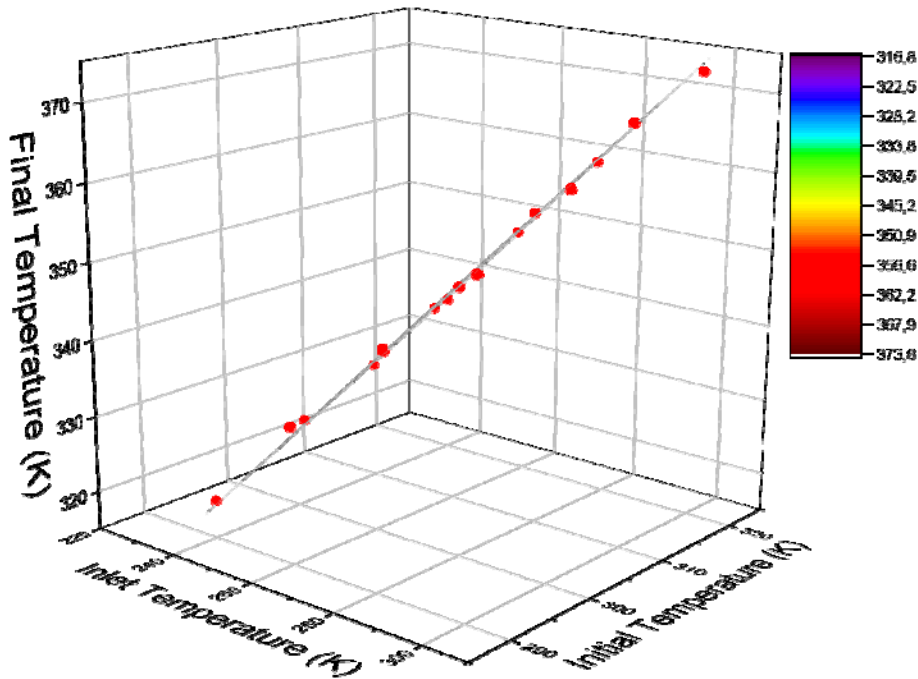


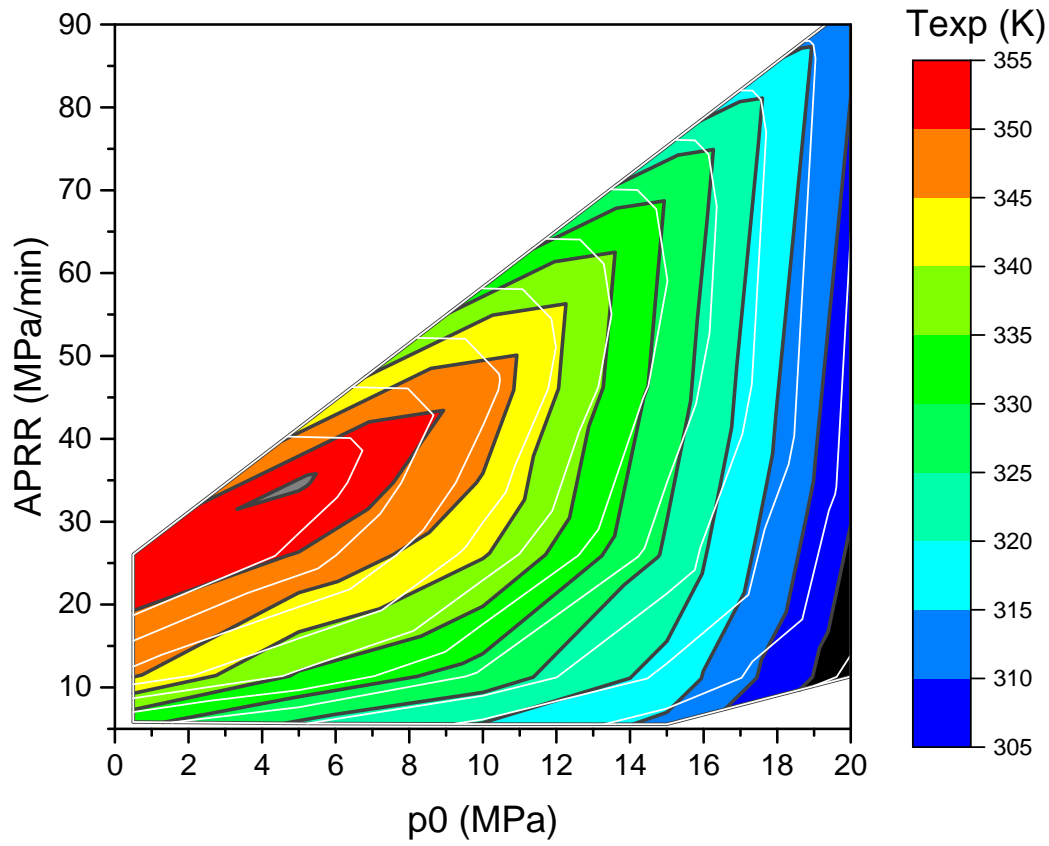
Figure 2. Fitted plane and data points [7] of final temperature via inlet and initial temperatures

3.3 Effect of initial pressure and average pressure ramp rate (APRR)

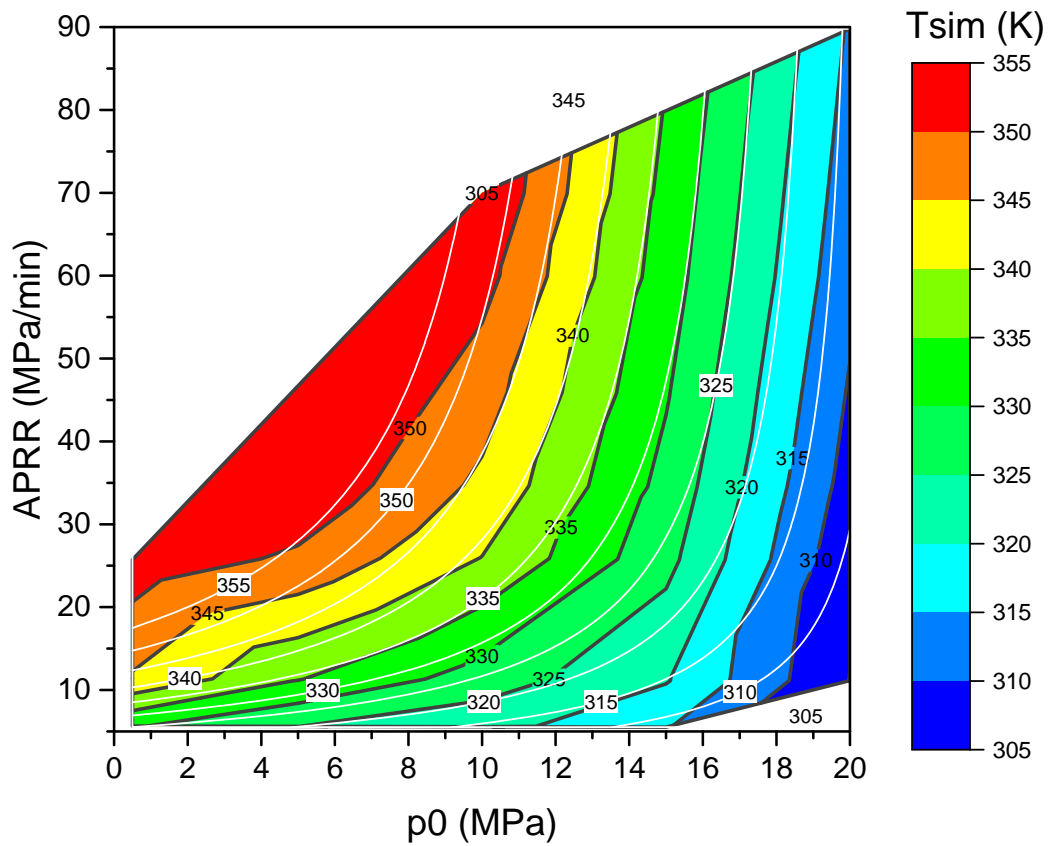
For charging process at room (ambient) temperature, $T_\infty = T_f$, the final hydrogen temperature function Eq.(7) is simplified to

$$T = \mu' T_0 + (1 - \mu') \frac{\gamma + \alpha}{1 + \alpha} T_f \quad (17)$$

Applying ideal gas of equation to initial and final status of the tank with volume V , we have $p_0 V = m_0 R T_0$ and $p V = m R T$, then we obtain $\mu = m_0 / m = (p_0 / p) (T / T_0)$. The time derivative of $p V = m R T$ can be written as $\dot{p} V = \dot{m} R T + m R \dot{T}$. During the charging process, the pressure change is mainly caused by mass flow rate rather than temperature change in the tank, so we can have $\mu = C(p_0 / p)$, where $C = T / T_0$ is assumed as a constant. For current experiments with constant final pressure p , we can further have $\mu = A p_0$. For simplification, we use $\mu' \approx \mu$. So we assume the modified initial mass fraction is approximately proportional to the initial pressure, i.e., $\mu' = A p_0$.



(a)



(b)

Figure 3. Contour comparison of fitted temperatures (white curves) with (a) experimental and (b) simulated temperatures (black curves [2]) in the plane of initial pressure and APRR

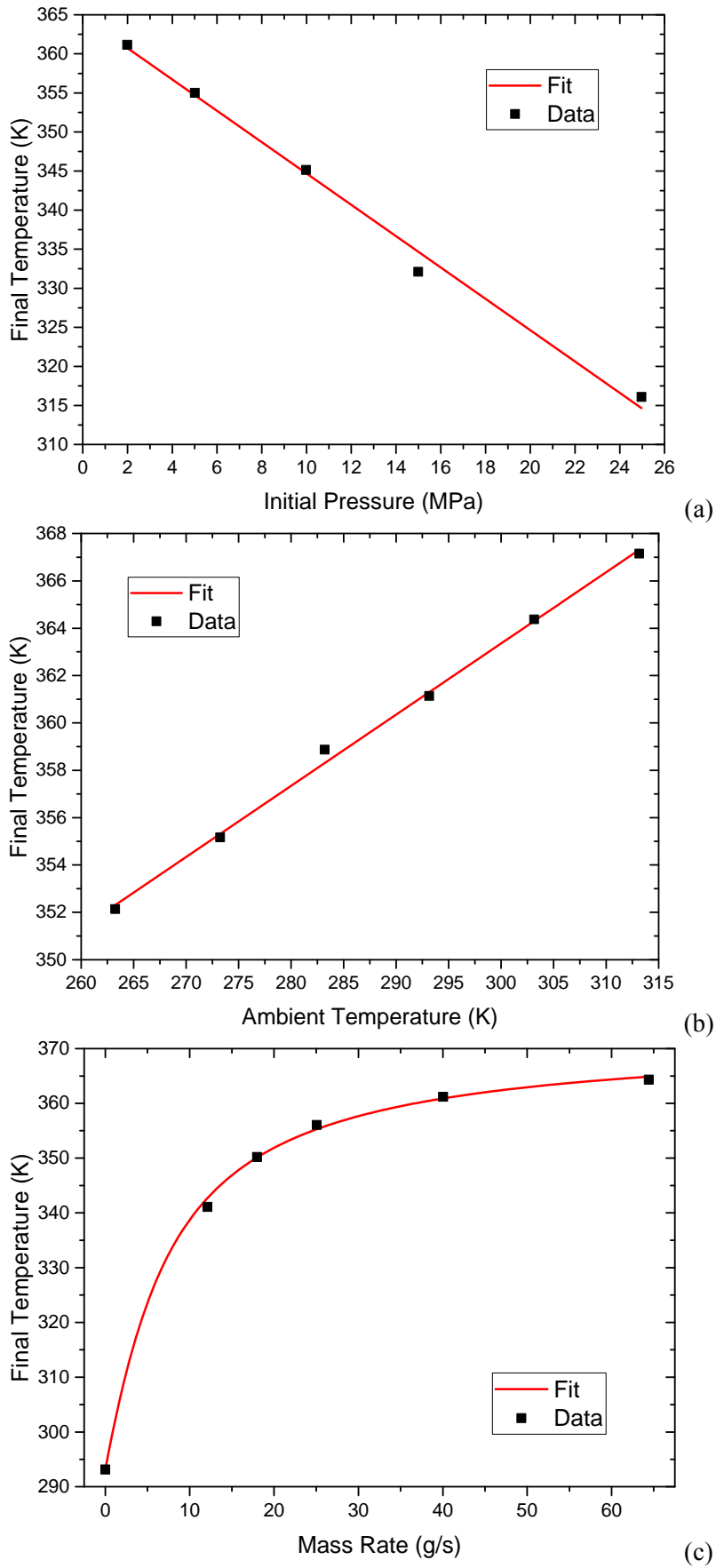


Figure 4. Effects of (a) initial pressure, (b) ambient temperature, (c) hydrogen mass flow rate on final hydrogen temperature compared with data [10]

From the definition $\alpha = (a_f A_s)/(c_v \dot{m})$, we can reasonably assume that the coefficient α is *inversely* proportional to the pressure change \dot{p} or average pressure ramp rate APRR approximately, i.e., $\alpha = B/\dot{p} = B/\text{APRR}$. The final hydrogen temperature function is

$$T = Ap_0 T_0 + (1 - Ap_0) \frac{\gamma + B/\dot{p}}{1 + B/\dot{p}} T_f \quad (18)$$

We fix $\gamma = 1.4$ and use $T_f = 293.15\text{K}$. In the current case, the inlet and initial temperatures are equal to the ambient temperature, i.e., $T_\infty = T_f$, $T_0 = T_f$. Therefore, the final hydrogen temperature is only the function of initial pressure p_0 and APRR. For the experimental and simulated data [2], the values of the fit parameters are shown in Table 4 and the results are shown in Figure 3. The fitting based on the simulated data is better because it has more data points.

Table 4. Fitted parameters of experimental and simulated temperature with initial pressure and APRR

Parameter		Value	Standard Error
Experimental temperature	A	0.04333	0.00125
	B	18.17276	1.26145
Simulated temperature	A	0.04192	8.49706E-4
	B	18.43238	1.14918

3.4 Effect of initial pressure, ambient temperature and mass flow rate

Substituting Eq.(4) into Eq.(7), the final hydrogen temperature becomes

$$T = \mu' T_0 + (1 - \mu') \frac{\gamma T_\infty + \alpha T_f}{1 + \alpha} \quad (19)$$

Experimental and simulated data are used to investigate the effects of initial pressure, ambient temperature and mass flow rate on final hydrogen temperature [10]. They agree well each other. We used the simulated data to fit Eq.(2) because it had more data points than available from the experimental data. Similar to Section 3.3, we assume $\mu' = K_p p_0$ and $\alpha = K_m / \dot{m}$, and fix $\gamma = 1.4$.

We use $T_0 = 293.15\text{K}$, $T_\infty = T_0$, and $T_f = T_0$ except in the case of variable ambient temperature. For the simulated data [10], the fitted parameters are shown in Table 5, the fitted results are shown in Figure 4. The fittings agree very well. The factor $(1 - \mu')\alpha/(1 + \alpha) = 0.30067$ for the ambient temperature agrees with the number of 0.3 from linear fitting [10].

Table 5. Effects of initial pressure, ambient temperature and mass flow rate

Effect	Parameter	Value	Standard Error	Note
Initial pressure p_0	Kp	0.02801	9.63398E-4	$\mu' = K_p p_0$
	alpha	0.63778	0.03071	α
Ambient temperature T_f	mu	0.24475	0.00832	$\mu' = \mu^{1+\alpha}$
	alpha	0.51736	0.01312	α
Mass flow rate \dot{m}	mu	0.32606	0.00785	$\mu' = \mu^{1+\alpha}$
	Km	13.98119	0.6466	$\alpha = K_m / \dot{m}$

4 CONCLUSIONS

(1) The rule of mixtures is a general weighted average method, which has been widely used to estimate various properties of a composite materials, porous media and multiphase system. The effective (moderate) temperature of the mixture (cold and warm hydrogen, or hydrogen and tank wall,

or even hydrogen and porous adsorbent) can be estimated based on the energy balance method. The energy, like density and heat capacity, is not sensitive with the structure of the mixture. Therefore, the *upper bound* of the rule of mixtures is used to estimate the hydrogen temperature.

(2) Inspired by the concept of the rule of mixture, efforts have been made to estimate effective/equivalent values of *physical state variables*, such as stresses in the composite materials or temperature in a system with mixtures. The analytical solution of hydrogen temperature behaves in an analogous way as the rule of mixtures. The final hydrogen temperature is the weighted average of initial temperature and a characteristic temperature which is related the inflow enthalpy of hydrogen. The weighted factor is initial heat capacity fraction.

(3) The simple uniform formula, inspired by the concept of the rule of mixtures with its weighted factors obtained from the analytical solution of thermodynamic model, is applied to fit published experimental or simulated results. These results show effect of initial and final mass, effect of inlet and initial temperatures, effect of initial pressure and average pressure ramp rate (APRR), effect of initial pressure, ambient temperature and mass flow rate. The fittings agree very well with the original data.

ACKNOWLEDGMENTS

We wish to thank the National Natural Science Foundation of China (NSFC Project No.51476120) and the Natural Sciences and Engineering Research Council of Canada (NSERC) for their financial support and Mr. Benjamin Angers for his technical assistance on the OriginPro 2015 software.

APPENDIX A. SOLUTIONS OF MASS AND TEMPERATURE IN CHARGE AND DISCHARGE PROCESSES

Mass and energy balance equations for the hydrogen gas stored in a tank are mostly written as

$$\frac{dm}{dt} = \dot{m}_{in} - \dot{m}_{out} \quad (A1)$$

$$\frac{d}{dt}(mu) = \dot{m}_{in} h_{in} - \dot{m}_{out} h_{out} + \dot{Q} \quad (A2)$$

where \dot{m}_{in} and \dot{m}_{out} are the hydrogen mass flow rates in which the hydrogen flows into the inlet and out of the exit of the tank respectively. Let us define \dot{m} as an algebraic quantity which can be positive or negative, say, it is positive for charge process, $\dot{m} = \dot{m}_{in}$, but it is negative for discharge process, $\dot{m} = -\dot{m}_{out}$. Further we have $\dot{m}h = \dot{m}_{in}h_{in}$ for charge process, $\dot{m}h = -\dot{m}_{out}h_{out}$ for discharge process. Thus, the above equations can be written as

$$\frac{dm}{dt} = \dot{m} \quad (A3)$$

$$\frac{d}{dt}(mu) = \dot{m}h + \dot{Q} \quad (A4)$$

Solution of mass balance equation (A3) under constant charge or discharge rate \dot{m} is simply a linear function of time:

$$m = m_0 + \dot{m}t \quad (A5)$$

Expanding the term of left hand side of the energy balance equation (A4) and utilizing mass solution (A5), the energy balance equation becomes

$$(m_0 + \dot{m}t) \frac{du}{dt} + \dot{m}u = \dot{m}h + \dot{Q} \quad (A6)$$

Divided by mass flow rate \dot{m} , we obtain energy balance equation in the form of specific internal energy u (J/kg):

$$(t^* + t) \frac{du}{dt} + u = h + q \quad (A7)$$

where $t^* = m_0 / \dot{m}$ is a characteristic time, the specific heat inflow q (J/kg) is:

$$q = \frac{\dot{Q}}{\dot{m}} = \frac{a_f A_s}{\dot{m}} (T_f - T) =: c_v \alpha (T_f - T) \quad (\text{A8})$$

is similar to specific internal energy u or specific enthalpy inflow h , where $\alpha = (a_f A_s) / (c_v \dot{m})$ is a dimensionless heat transfer coefficient which represents the ratio of *heat transfer ability* $a_f A_s$ to *total heat capacity change* $c_v \dot{m}$ (W/K) of the system during charge and discharge processes. For ideal gas with constant specific heats, we can have the specific internal energy $u = c_v T$ and specific enthalpy $h = c_p T$, and we use the notation $\gamma = c_p / c_v$. For the case of constant inflow or outflow hydrogen temperature T_∞ , we denote the specific enthalpy of either inflow or outflow hydrogen as $h_\infty = c_p T_\infty$.

With these assumptions and notations, the energy equation (A7) can be written as

$$\frac{dT}{dt} = (1 + \alpha) \frac{T^* - T}{t^* + t}, \quad T^* = \frac{\gamma T_\infty + \alpha T_f}{1 + \alpha} \quad (\text{A9})$$

where T^* is a characteristic temperature. Using dimensionless time $\tau = t / t^*$, the solution of equation (A9) can be obtained by integration as:

$$\frac{T^* - T}{T^* - T_0} = \left(\frac{1}{1 + \tau} \right)^{1 + \alpha} \quad (\text{A10})$$

REFERENCES

1. Dicken, C.J.B. and Mérida, W., Measured effects of filling time and initial mass on the temperature distribution within a hydrogen cylinder during refuelling, *Journal of Power Sources*, **165**, 2007, pp. 324-336.
2. Striednig, M., Brandstatter, S., Sartory, M. and Klell, M., Thermodynamic real gas analysis of a tank filling process, *International Journal of Hydrogen Energy*, **39**, 2014, pp. 8495-8509.
3. Zheng, J.Y., Guo, J.X., Yang, J., Zhao, Y.Z., Zhao, L., Pan, X.M., Ma, J.X. and Zhan, L.F., Experimental and numerical study on temperature rise within a 70 MPa type III cylinder during fast refuelling, *International Journal of Hydrogen Energy*, **38**, 2013, pp. 10956-10962.
4. Guo, J.X., Yang, J., Zhao, Y.Z., Pan, X.M., Zhang, L.F., Zhao, L. and Zheng, J.Y., Investigations on temperature variation within a type III cylinder during the hydrogen gas cycling test, *International Journal of Hydrogen Energy*, **39**, 2014, pp. 13926-13934.
5. Ortiz Cebolla, R., Acosta, B., Moretto, P., Frischauf, N., Harskamp F., Bonato C. and Baraldi D., Hydrogen tank first filling experiments at the JRC-IET GasTeF facility, *International Journal of Hydrogen Energy*, **39**, 2014, pp. 6261-6267.
6. Acosta, B., Moretto, P., de Miguel, N., Ortiz, R., Harskamp, F. and Bonato, C., JRC reference data from experiments of on-board hydrogen tanks fast filling. *International Journal of Hydrogen Energy*, **39**, 2014, pp. 20531-20537.
7. de Miguel, N., Acosta, B., Baraldi, D., Melideo, D., Ortiz, R. and Moretto, P., Experimental and numerical analysis of refueling of on-board hydrogen tanks at different ambient temperatures. The 20th World Hydrogen Energy Conference (WHEC2014), Gwangju, Korea, June 15-20, 2014.
8. de Miguel, N., Ortiz Cebolla, R., Acosta, B., Moretto, P., Harskamp, F. and Bonato, C., Compressed hydrogen tanks for on-board application: Thermal behaviour during cycling, *International Journal of Hydrogen Energy*, **40**, 2015, pp. 6449-6458.
9. Ortiz Cebolla, R., Acosta, B., de Miguel, N. and Moretto, P., Effect of precooled inlet gas temperature and mass flow rate on final state of charge during hydrogen vehicle refuelling, *International Journal of Hydrogen Energy*, **40**, 2015, pp. 4698-4706.
10. Zhao, L., Liu, Y.L., Yang, J., Zhao, Y.Z., Zheng, J.Y., Bie, H.Y. and Liu, X.X., Numerical simulation of temperature rise within hydrogen vehicle cylinder during refueling, *International Journal of Hydrogen Energy*, **35**, 2010, pp. 8092-8100.

11. Zhao, Y.Z., Liu, G.S., Liu, Y.L., Zheng, J.Y., Chen, Y.C., Zhao, L., Guo, J.X. and He, Y.T., Numerical study on fast filling of 70 MPa type III cylinder for hydrogen vehicle, *International Journal of Hydrogen Energy*, **37**, 2012, pp. 17517-17522.
12. Wang, L., Zheng, C.X., Li, R., Chen, B.B. and Wei, Z.X., Numerical analysis of temperature rise within 70 MPa composite hydrogen vehicle cylinder during fast refueling, *Journal of Central South University*, **21**, 2014, 2772-2778.
13. Heitsch, M., Baraldi, D. and Moretto, P., Numerical investigations on the fast filling of hydrogen tanks, *International Journal of Hydrogen Energy*, **36**, 2011, pp. 2606-2612.
14. Galassi, M.C., Baraldi, D., Acosta Iborra, B. and Moretto, P., CFD analysis of fast filling scenarios for 70MPa hydrogen type IV tanks, *International Journal of Hydrogen Energy*, **37**, 2012, pp. 6886-6892.
15. Melideo, D., Baraldi, D., Galassi, MC, Ortiz Cebolla, R, Acosta Iborra, B and Moretto, P., CFD model performance benchmark of fast filling simulations of hydrogen tanks with pre-cooling. *International Journal of Hydrogen Energy*, **39**, 2014, pp. 4389-4395.
16. Galassi, M.C., Papanikolaou, E., Heitsch, M., Baraldi, D., Iborra, B.A. and Moretto P., Assessment of CFD models for hydrogen fast filling simulations, *International Journal of Hydrogen Energy*, **39**, 2014, pp. 6252-6260.
17. Ahluwalia, R.K., Hua, T.Q. and Peng, J.K., On-board and off-board performance of hydrogen storage options for light-duty vehicles, *International Journal of Hydrogen Energy*, **37**, 2012, pp. 2891-2910.
18. Yang, J.C. and Huber, M.L., Analysis of thermodynamic processes involving hydrogen, *International Journal of Hydrogen Energy*, **33**, 2008, pp. 4413-4418.
19. Yang, J.C., A thermodynamic analysis of refueling of a hydrogen tank. *International Journal of Hydrogen Energy*, **34**, 2009, pp. 6712-6721.
20. Ahluwalia, R.K. and Peng, J.K., Dynamics of cryogenic hydrogen storage in insulated pressure vessels for automotive applications. *International Journal of Hydrogen Energy*, **33**, 2008, pp. 4622-4633.
21. Ahluwalia, R.K. and Peng, J.K., Automotive hydrogen storage system using cryo-adsorption on activated carbon. *International Journal of Hydrogen Energy*, **34**, 2009, pp. 5476-5487.
22. Kumar, V.S., Raghunathan. K. and Kumar, S., A lumped parameter model for cryo-adsorber hydrogen storage tank. *International Journal of Hydrogen Energy*, **34**, 2009, pp. 5466-5475.
23. Xiao, J.S., Zhou, Z.Q., Cossement, D., Bénard, P. and Chahine, R., Lumped parameter model for charge-discharge cycle of adsorptive hydrogen storage system. *International Journal of Heat and Mass Transfer*, **64**, 2013, pp. 245-253.
24. Xiao, J.S., Li, Q., Cossement, D., Bénard, P. and Chahine, R., Lumped parameter simulation for charge-discharge cycle of cryo-adsorptive hydrogen storage system. *International Journal of Hydrogen Energy*, **37**, 2012, pp. 13400-13408.
25. Xiao, J.S., Bénard, P. and Chahine, R., Thermodynamic models of hydrogen storage systems. The 5th World Hydrogen Technology Convention (WHTC2013), Shanghai, China, September 25-28, 2013.
26. Xiao, J.S., Bénard, P. and Chahine, R., Adsorption-desorption cycle thermodynamics for adsorptive hydrogen storage system. The 20th World Hydrogen Energy Conference, Gwangju, Korea, June 15-20, 2014.
27. Xiao, J.S., Bénard, P. and Chahine, R., Estimation of final gas temperature in compressed hydrogen vehicle refuelling. The 1st International Conference on Modern Auto Technology and Services (ICMATS2014), Wuhan, China, October 24-26, 2014.
28. Xiao, J.S., Zhan, Z.G., Zhao, W.H. and Fu, W.B., Lower and upper bounds of mechanical and thermal properties of ceramic/metal composites, *Key Engineering Materials*, **336-338**, 2007, pp. 2440-2443.
29. Kim, H.S., Hong, S.I. and Kim, S.J., On the rule of mixtures for predicting the mechanical properties of composites with homogeneously distributed soft and hard particles. *Journal of Materials Processing Technology*, **112**, 2001, pp. 109-113.