



Modelling activated carbon hydrogen storage tanks using machine learning models



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ABSTRACT

The application of hydrogen for energy storage and as a vehicle fuel necessitates efficient and effective storage technologies. In addition to traditional cryogenic and high-pressure tanks, an alternative approach involves utilizing porous materials such as activated carbons within the storage tank. The adsorption behaviour of hydrogen in porous structures is described using the Dubinin-Astakhov isotherm. To model the flow of hydrogen within the tank, we rely on the equations of mass conservation, the Navier-Stokes equations, and the equation of energy conservation, which are implemented in a computational fluid dynamics code and additional terms account for the amount of hydrogen involved in sorption and the corresponding heat release. While physical models are valuable, data-driven models often offer computational advantages. Based on the data from the physical adsorption model, a data-driven model is derived using various machine learning techniques. This model is then incorporated as source terms in the governing conservation equations, resulting in a novel hybrid formulation which is computationally more efficient. Consequently, a new method is presented to compute the temperature and concentration distribution during the charging and discharging of hydrogen tanks and identifying any limiting phenomena more easily.

1. Introduction

1.1. Hydrogen storage

With the transition away from fossil fuels in the energy sector and the advancement of renewable energy sources, the significance of hydrogen as a means of storing and transporting energy is steadily growing. Hydrogen, stored as a fuel for trucks and trains, as well as a seasonal storage solution for excess electricity generated by renewables, plays a crucial role. Moreover, there is a vision for hydrogen to replace a significant portion of the currently used fossil fuels. Consequently, the efficient storage of hydrogen has become paramount.

The storage of gases, particularly hydrogen, poses challenges primarily due to the low density of hydrogen. Despite its ability to store a substantial amount of energy relative to its mass (gravimetric energy density), the volume-specific energy storage (volumetric energy density) of hydrogen is comparatively minimal.

To minimize the required storage volume for hydrogen, various methods are employed, including liquefaction at extremely low temperatures (liquid hydrogen, LH₂), compression at high pressure (compressed hydrogen, CH₂), and a combination of low temperature with

high pressure (cryo-compressed hydrogen, CCH₂). Trucks utilize CH₂ tanks at pressures of 350 bar and 700 bar, while LH₂ tanks undergo field testing. Given the pivotal role of hydrogen in the transition to renewable energy sources, there are specific targets for enhancing the storage capacity of hydrogen tanks and tank systems, as depicted in Fig. 1. In addition to volumetric and gravimetric capacities, targets are also set for the charging and discharging rates, i.e. a system fill time 3–5 min and discharge rates up to 1 g/s, for heavy duty vehicles a charging flow rate of 10 g/min is envisaged. Achieving these targets necessitates substantial ongoing research and development efforts.

Storage at high pressure and low temperature incurs significant costs associated with equipment and energy losses during compression and cooling. An alternative approach involves the storage of hydrogen in solids through adsorption or chemisorption. Extensive efforts are underway to identify suitable storage materials and reduce the pressure and temperature differences required for storing a specific quantity of hydrogen.

In physical adsorption, gas molecules adhere to the surface of the adsorbent medium. Therefore, materials with high surface area and porosity are advantageous, leading to a higher density in the adsorbed phase compared to the gaseous phase. The adsorption process is

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exothermic. Effective adsorbents exhibit high porosity and inner surface area, such as activated carbons or metal-organic frameworks. These adsorbents are available in the form of powder, custom-shaped monoliths, or pellets, which are inserted into tanks.

During tank charging, the heat of adsorption may cause the adsorbent to heat up, while discharging results in cooling, influencing the adsorption equilibrium. These thermal effects are more pronounced in chemisorption.

For metal hydrides these effects are significant also at low hydrogen concentrations. For other materials, these effects get significant at the concentrations levels envisaged by the DOE targets. Thus appropriate methods are needed to efficiently and effectively describe these phenomena.

As the tank pressure rises, the quantity of hydrogen stored in the tank also increases, as illustrated in Fig. 2. A portion of the hydrogen undergoes adsorption, while the remaining portion remains in the gas phase as compressed gas within the voids of the porous adsorbent. With increasing pressure, the fraction of adsorbed hydrogen decreases. Therefore, the use of adsorbents for gas storage is more favorable at lower pressures, as the additional amount stored in a given volume compared to the gaseous state is higher. The optimal operational conditions hinge on the material properties of the gas and sorbent.

Hydrogen sorption tanks find applications in domestic power systems and small vehicles, prompting extensive research into the development of suitable sorbent materials and the optimization of tanking systems.

The application taken into consideration for this study is the storage of hydrogen as a vehicle fuel. Hydrogen tanks for vehicles comply to safety regulations with regard to leakage and permeation. For a 33 L tank the leakage rate is below $9 \cdot 10^{-3}$ mbar/l s (ISO/TS 15869), thus not affecting the pressure levels with regard to the pressure range and time range in this study. According to ISO 14687-2 (purity of hydrogen for fuel-cell vehicles) the water vapor content in hydrogen is lower than 5 $\mu\text{mol/mol}$. Thus for the pressure and temperature range in this analysis the water stays solved and no ice formation takes place. Accordingly also the influence of the other impurities is neglected in this analysis.

Lee et al. [1] provide a comprehensive review of the progress in adsorbent material development, considering the possibilities for

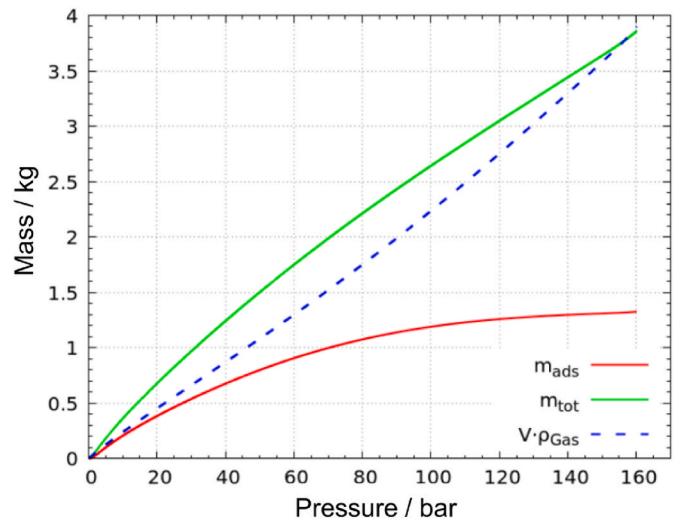


Fig. 2. Charging of a 33 L tank of activated carbon with hydrogen (293 K and 100 kPa at inlet). Stored mass depending on tank pressure. The total mass adsorbed (green line) increases as the pressure in the tank increases. The mass adsorbed (red line) increases as the pressure increases but significantly more reduced at higher pressure. The difference is the mass of hydrogen compresses in the voids of the porous media. With tank pressure higher than 160 bar there is no additional amount of hydrogen stored compared to a simple compression tank.

realization and commercialization.

Mohan et al. [2] outline the state of the art using carbon materials (activated carbon, graphite, carbon nanotubes, and fibers) as adsorbents, along with advantageous production methods and tank systems.

Shet et al. [3] discuss the latest strategies to enhance the surface area of Metal-Organic Frameworks (MOFs) and improve hydrogen uptake.

A technical comparative analysis [4] of the different physical and material based types of hydrogen storage systems illustrates the paradoxical inherent features, including gravimetric and volumetric storage densities and parameters associated with storage and release processes,

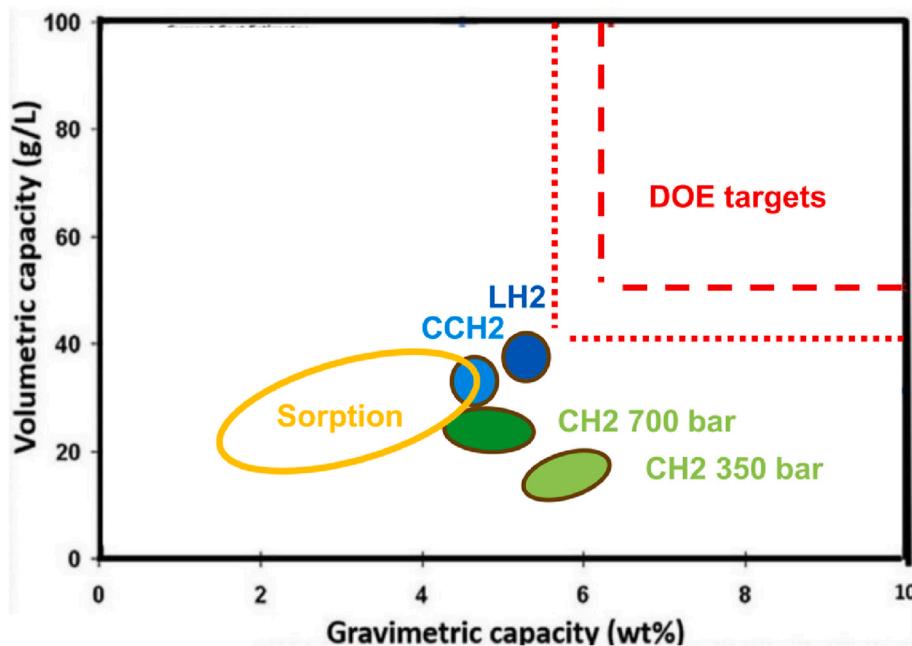


Fig. 1. Volumetric storage capacity and gravimetric storage capacity for storage tanks with compressed hydrogen (CH2), liquefied hydrogen (LH2), cryo-compressed hydrogen (CCH2), sorption storage and US Department of Energy (DOE) targets.

among these systems. Accordingly, no ideal hydrogen storage technique can be considered the best-fit for all stationary and automotive applications. Therefore the authors conclude that not only a unique hydrogen storage solution can properly provide the needs, but a set of complementary solutions which may offer the system designer several options.

Meduri et al. [5] provide an overview on materials suitable for storing hydrogen at room temperature. The critical issues and challenges in developing able materials for hydrogen storage at room temperature is discussed and various techniques to improve the hydrogen uptake at room temperature are presented.

Chen et al. [6] highlight the progress related to porous-material-based composites with encapsulated hydrides and metal organic frameworks, the engineering of materials into practical gas vessels as well as future commercialization. Porous materials are a viable technology for hydrogen storage, nevertheless materials with improved gravimetric and volumetric storage capacities at ambient temperatures are needed.

An analysis of operational challenges and recent advancements in hydrogen storage techniques is presented by Bosu et al. [7]. The comparative performance of various methods is tabulated and the existing research gap and future perspectives are analysed.

In order to facilitate this ongoing research, to optimize the adsorbent and tank shapes, the temperature control and operating conditions, effective and efficient modelling methods describing the sorption storage are needed.

1.2. Modelling hydrogen adsorption storage

For specific materials and operating conditions where the temperature, pressure, and adsorbed phase distribution in the adsorbent and tank are almost uniform, a lumped parameter model can be employed. However, when dealing with high mass flux during charging and discharging, a dense adsorbent (as in certain monoliths), or the introduction of additional heating or cooling measures, the homogeneity of property values and concentration is compromised. To describe these non-uniform phenomena, a spatially resolved model becomes necessary.

The flow within porous media is intricately linked to heat and mass transfer resulting from the adsorption process. The operational approach involves either using a lumped parameter model that overlooks spatial distribution and fluid flow or solving the heat transport equations neglecting the flow or employing a Computational Fluid Dynamics (CFD) code for flow computation as a foundation and integrating additional physical model equations describing adsorption. This implementation is labour-intensive. Furthermore, if gas mixture equilibrium or chemical reactions need to be computed, the additional iterations may significantly increase computation time.

In the pursuit of optimizing tank and tanking system design, a method that efficiently describes and predicts physisorption and chemisorption is crucial. While lumped parameter models are efficient, they fall short in predicting the non-uniform temperature distribution in tanks, which holds significance for safety considerations.

1.2.1. Lumped parameter model

Lumped parameter models are very good at predicting the charging and discharging for a homogeneous temperature and concentration distribution in the sorbent, nevertheless if the distribution is uneven the results are to inaccurate and key features like hot spots get undetected.

An integrated model of a sorbent-based cryogenic compressed hydrogen system is used to assess the prospect of meeting the DOE targets for hydrogen-fuelled vehicles [8]. The model includes the thermodynamics of sorption, heat transfer during adsorption and desorption, sorption dynamics, energetics of cryogenic tank cooling, and containment of hydrogen in wound carbon fibre tanks.

Thermal affects during charging of hydrogen in a 2 L tank with activated carbon at 300 K are investigated [9]. Depending on the adsorption capacity at 300 K, the temperature increase that takes place

during the charging process is either mainly due to the conversion to heat of the work of the compression force (capacities smaller 2 wt%) or due to adsorption (towards capacities of 6 wt%).

System simulation models for a cryo-adsorption system and a metal hydride systems are performed based on lumped parameter models [10]. Simulation results to test the storage systems' ability to meet fuel cell demand for different drive cycles and varying operating conditions are presented and compared to the DOE system goals.

A low-cost lumped parameter model is developed to simulate hydrogen storage in solid-state materials. It is capable of predicting key thermofluidic quantities of the storage system, such as the storage bed temperature and pressure, hydrogen fraction and flowrate, and storage tank temperature with reasonable accuracy at much lower costs than a CFD model [11].

1.2.2. Heat transfer model

In order to assess the need for cooling and thermal stresses the heat of adsorption and heat transfer to the ambient may be taken into account. Nevertheless in this way the effect of flow conditions and concentration variations cannot be considered appropriately.

Hydrogen absorption-desorption in a metal hydride bed reactor is modelled [12] and the diffusion and heating of hydrogen and metal hydride powder in both radial and axial directions is simulated. The model consists of system of partial differential equations (PDE) describing two-dimensional heat and mass transfer of hydrogen in a porous matrix. The influence of the operating parameters temperature, pressure, concentration, permeability and thermal conductivity on the rate of absorption-desorption of hydrogen in metal hydride is discussed.

A portable hydrogen tank using adsorption material is designed using FEM for stress analysis and heat transport. Here the fluid behaviour is not addressed, the heat of adsorption and desorption for various adsorbents is taken into account [13].

1.2.3. CFD models

The most comprehensive approach is to consider the heat transfer, the flow and the sorption process. This is mostly achieved by incorporating the physical model describing the adsorption into a Computational Fluid Dynamics (CFD) code.

Ye et al. [14] implemented the Dubinin-Astakhov adsorption isotherm into a Computational Fluid Dynamics (CFD) code to calculate mass and energy source terms. They compared the computed results with measured values for the charging and discharging of a 2.4-L hydrogen tank at ambient conditions up to 10 MPa, using activated carbon as the adsorbent.

A CFD model was employed by Xiao [15] to simulate the charge-discharge cycle of hydrogen storage adsorption on activated carbon at cryogenic temperatures, aiming to optimize the system. Additionally, Xiao [16] simulated and compared charge-discharge cycles of two MOF-5 adsorbents with activated carbon.

Chung et al. [17] used a similar approach to model hydrogen storage in MOFs, comparing different tank designs and heat transfer enhancement strategies.

A tempered metal hydride thermal energy storage reactor design was optimized by Bao [18], evaluated through a 3D CFD simulation. The study investigated parameters like the number of heat exchanger tubes, tube diameter, and tube pitch, aiming to maximize the average heat storage rate per unit mass.

Ubaid [19] numerically and experimentally investigated the charging of hydrogen tanks with MOFs as adsorbents at low (77 K) and ambient temperatures and pressures up to 9 MPa. The study included results for activated carbon for comparison.

Xu [20] explored the impacts of various factors on cryogenic hydrogen storage in MOFs, including mass flow rate, outlet opening time, bed density, and heating power

Ortmann [21] modelled the refuelling of a single MOF-5 pellet with cryogenic hydrogen at moderate pressures, studying the variation in

refuelling time with different pellet sizes.

Banerjee [22] compared different approaches to compute coefficients and property values of a CFD model, validating the data with Ye's results. Furthermore, Bannejee presented results from measurements and simulations of hydrogen adsorption in a novel activated carbon at ambient temperatures and pressures up to 10 MPa.

Chibani [23–25] assessed the performance of a large-scale metal-hydride storage reactor with metal foam-phase change material tubes and a large-scale activated carbon bottle surrounded by a layer of metal foam using 2D transient numerical models. The study analysed various materials and geometries to assess the potential of phase change materials in reducing hydrogen storage costs.

Ebne [26] developed and validated a CFD model for simulating the refuelling process throughout the entire equipment of a hydrogen refuelling station. The model accurately replicated hydrogen temperature and pressure, enabling the development of fuelling protocols for arbitrary parameters, although computational costs remain significant.

These models are elaborate and describe the relevant phenomena. Nevertheless due to the incorporation of the equations describing the adsorption necessitating additional iterations and even restricting the maximum time step the computation is expensive. Thus the resulting studies are limited to an assessment or comparison of basic design alternatives.

1.2.4. Machine learning models

With machine learning (ML), algorithms are developed which learn from data without any explicit physical model. Lately there have been significant advances with regard to the methods available and their successful application.

Machine Learning (ML) is employed to enhance Computational Fluid Dynamics (CFD) through the development of advanced reduced-order models, as demonstrated by Vinuesa [27]. One main target of these efforts is to accelerate the performance of CFD models, enhancing them with ML methods. These are used to model complex physical phenomena in a computational more advantageous form. This approach is utilized here to streamline the model describing the adsorption process, subsequently integrating it into the CFD code.

As an alternative to measurements and physical modelling of the sorption process, machine learning models are applied in order to achieve an efficient high throughput screening of potential sorbents. Thus the sorption process is described without specifically modelling the internal structure and reactions. Research by Borja [28], Jie [29], and Klepp [30] indicates that ML models can effectively predict the gravimetric and volumetric hydrogen uptakes of porous structures. As methods linear and non-linear regression, decision trees, artificial neural networks. Notably, various formulations of artificial neural networks (NN) and Gaussian Process Regression (GPR) yield the most accurate predictions for volumetric and gravimetric uptake in metal-organic frameworks [28],

GPR is a probabilistic model that defines a distribution over all possible functions consistent with the data and prior assumptions. It is particularly suitable for small datasets, offering flexibility in representation and inherent uncertainty measures over predictions. Gaussian processes assume the output variable is a realization of a Gaussian process, with a prior distribution specified and probabilities adjusted based on evidence (observed data). The chosen mean function and covariance kernel function are crucial aspects of GPR. The selected mean function here is constant (mean of the training dataset), and the exponential and squared exponential kernels yield optimal results.

In contrast, NN consist of layers of interconnected artificial neurons with adjustable weights and biases, learning optimal values by minimizing a loss function on a training dataset. While NN is a deterministic model with sparse matrix solvers, GPR provide uncertainty estimates and adapt to different data types through the choice of an appropriate kernel function. However, for very large datasets, the computational requirements of GPR can become prohibitive.

Here we assess the potential of NN and GPR to successfully describe the adsorption process and use the resulting ML model to incorporate it into a CFD model.

1.3. Problem description

A hybrid approach, as an alternative to a purely physical model, involves replacing a portion of the physical model and its intricate equations with a data-driven model featuring simpler equations.

In the novel approach described here, a CFD code serves as the foundation and the sorption mechanism is implemented using a ML-model replacing the physical model. This choice aims to simplify both implementation and computation when modelling the charging and discharging of hydrogen in tanks filled with adsorbent.

As an example for this approach an analysis of hydrogen storage in carbon materials by physisorption is performed, as there is sufficient experimental data available and it is of significant importance with regard to the energy transition in the mobility sector. This approach can easily be extended to other adsorbates, adsorbent materials and chemisorption.

The paper is organized as follows: The next section (Section 2 Methods) gives the physical equations (conservation equations and adsorption model) used to model the problem and describes the machine learning (ML) approach used. The ML model with the best performance is incorporated as source term into a CFD code. The results are compared to experimental data and data generated by the physical model. Some results obtained from this hybrid model are shown and discussed in Section 3. We highlight strategies to overcome limitations of the storage capacity of adsorbent tanks that use adsorbent packed beds in order to meet the DOE recommendation. Finally an outlook of the implications of the results obtained and the method described here is given.

This study has some new features: The adsorption of hydrogen in carbon materials is modelled using several ML algorithms to predict adsorbed mass and heat flux. A model based on the GPR algorithm is incorporated in a CFD code resulting in an efficient and effective hybrid model for the computation of the charging and discharging of hydrogen in carbon adsorption tanks. As this approach is used for the first time a number of issues pending further investigations arise. The approach presented here acts as a blue print for a more general approach modelling sorption for different materials and perspectively chemical reactions in a way convenient for the implementation in a CFD code.

2. Method

2.1. Hybrid models

For intricate physical problems such as the sorption of gases in porous beds, encompassing flow, heat and mass transfer, multiple phases and species, as well as reactions, numerical modeling becomes prohibitively expensive when dealing with a wide range of time and space scales. To address this challenge, hybrid models are proposed: a portion of the problem is delineated by fundamental physical equations, involving a set of coupled nonlinear partial differential equations, while other phenomena are described by simpler models, typically sets of algebraic equations. These simpler models are derived using machine learning methods based on datasets generated through measurements as well as simulations. The optimization of the methodology for deriving these data-driven models and coupling them with physical models remains an ongoing area of research. This work is a contribution to this goal.

At first the set of equations describing the problem is given. Then the machine learning approach is described: The datasets generated for training and validation, derived from the physical model are described as well as the input and output parameters chosen. Different approaches for the machine learning are used and the results with regard to accuracy and predictability are compared. A hybrid model for the charging and

discharging of adsorption tanks is generated by incorporating the ML adsorption model into a CFD code. Results using this data driven ML model are compared to experimental data as well as with results using a physical adsorption model.

2.2. Physical model

2.2.1. Adsorption model

The Dubinin-Astakhov equation for the adsorption isotherm is given by

$$q_i = \rho_{ads} W_0 \exp \left[- \left(\frac{A}{b E_0} \right)^n \right] \quad (1)$$

with the maximum mass of adsorbed gas $q_{max} = \rho_{ads} W_0$ given by the adsorbent density ρ_{ads} and the micropore volume of the adsorbent W_0 and the reference adsorption energy E_0 and the affinity coefficient for the adsorbent-adsorbate interaction b and the exponent n (close or equal to 2). The free energy of adsorption $E_0 = \alpha + \beta T$ is described as a function of temperature T , an enthalpic factor α and an entropic factor β , depending of the materials involved.

The adsorption potential $A = R T \ln(P_s/P)$ is determined with the saturated vapor pressure calculated from the critical values $P_s = P_{cr} (T/T_{cr})^2$.

According to Richard [31] for the supercritical conditions, as encountered for the adsorption of hydrogen at ambient temperature, a better agreement is achieved by fitting the maximum amount of adsorbed gas n_{max} .

The isosteric heat of adsorption for $n = 2$ is given by

$$H = \alpha \sqrt{q_{max}/q_i} \quad (2)$$

The adsorption rate may be computed by

$$dq/dt = k(q_i - q) \quad (3)$$

according to the linear driving force model [32] with the mass transfer coefficient at an aggregated level k .

Alternatively, in the case of slow charging rates the equilibrium absolute adsorption amount may be used for simplicity, and the adsorption rate is determined directly by differentiating the equation for q_i

2.2.2. Conservation equations

The flow dynamics within the tank's free space, unoccupied by adsorbent, is characterized by the Navier-Stokes equations, which are modified to account for the effects of the adsorbent material. For gaseous hydrogen, the mass balance equation incorporates adsorption and desorption processes through source terms. The flow passing through the adsorbent, treated as a porous medium, is described by the Darcy-Forchheimer equation. This equation introduces a resistance to the flow, effectively acting as a sink in the momentum balance equations, where it is included as a source term. In the energy balance, the heat retained by the solid adsorbent is accounted for by calculating an effective heat capacity, which integrates the heat capacity of the adsorbent itself. Furthermore, the heat generated or consumed by the adsorption process is incorporated as an additional source term, directly tied to the mass flux of adsorbed hydrogen.

The mass conservation equation is given by

$$\epsilon \frac{\partial \rho_g}{\partial t} + \vec{\nabla} \bullet (\rho_g \vec{V}) = S_m \quad (6)$$

with the velocity vector V and the mass source term

$$S_m = -(1 - \epsilon) \rho_p \frac{\partial q}{\partial t} \quad (7)$$

The momentum conservation equation is given by

$$\frac{\rho_g}{\epsilon} \left[\frac{\partial \vec{V}}{\partial t} + \vec{V} \bullet \vec{\nabla} \left(\frac{\vec{V}}{\epsilon} \right) \right] = -\vec{\nabla} \bullet P + \frac{\mu}{\epsilon \rho_g} (\Delta \vec{V}) + S_D \quad (8)$$

taking into account the flow in the porous bed

$$S_D = - \left[\frac{\mu}{K} + \frac{1.75 \rho_g}{\sqrt{150 K \epsilon^3}} |\vec{V}| \right] \vec{V} \quad (9)$$

The energy conservatio equation is given by

$$\begin{aligned} \frac{\partial \rho e}{\partial t} + \nabla \bullet (\rho \vec{V} e) + \frac{\partial \rho K}{\partial t} + \nabla \bullet (\rho V \vec{V} K) + \nabla \bullet (p \vec{V}) - \nabla \bullet (h_{eff} \nabla e) \\ = \rho u \bullet g + f_{rad}(e) + S_e \end{aligned} \quad (10)$$

With the internal energy e , the kinetic energy K and the transfer coefficient h_{eff} .

Gravitational energy and radiation energy transfer are neglected, in the energy conservation equation an additional source term accounts for the heat of adsorption

$$S_e = -S_m \frac{H}{M_{H_2}} \quad (11)$$

At the inlet and at the outlet, a velocity value is prescribed, derived from the pre-scribed mass flow rate. At the walls the heat exchange with the ambient is approximated with Newtons law.

The flow is computed with openFoam11 as a CFD code [33]. The flow is laminar and adjusted to the porosity of the adsorbent. The influence of adsorption and de-sorption is taken into account by coded source terms in the energy and mass conservation equation.

Property values for the adsorbate and the hydrogen are taken from literature [34,35], see Table 1.

2.3. Machine learning model

To develop the machine learning model for adsorption, a comprehensive dataset comprising 4500 entries for training and validation, and an additional dataset with 1500 entries for testing, has been compiled. These datasets simulate the adsorption of hydrogen in activated carbons with specific model constants outlined in Table 1.

2.3.1. Data set

To develop the machine learning model for adsorption, a comprehensive dataset comprising 4500 entries for training and validation, and an additional distinct data set with 1500 entries for testing, has been compiled. These datasets simulate the adsorption of hydrogen in activated carbon, with specific model constants outlined in Table 1.

The data sets are derived from the physical adsorption model based on the DA isotherm. Thus requirements on the data sets with regard to the distribution of the data points (e.g. range and spacing) is much more

Table 1

Model parameters for the adsorption of hydrogen in activated carbon used in this analysis. As base case data for generic activated carbon (AC) is used, in addition also a model for the high performance carbon AX-21 is generated.

ML method	AC	AX-21
Temperature Range [K]	250–350	50–350
Pressure Range [MPa]	0.1–40	0.1–70
Adsorption		
q_{max} g/kg	3.3	143
α J/mol	5508	3180
β J/mol K	–	18.9
n	2	2
Material Properties		
Density kg/m ³	500	269
Conductivity W/m K	0.650	0.764
Heat Capacity J/kg K	650	825
Bed porosity	0.65	0.49

easily met, then for measured data. In addition using the model data for a variety of different materials and operating conditions is easily computed. Using a model instead of measured data requires an additional validation of the ML model against measured data. The data is generated along charging and discharging cycles with variations in the initial temperature and the initial and final pressure.

The datasets span a pressure range from 0.1 to 10 MPa and a temperature range from 50 K to 350 K. Input variables to the model are temperature and pressure, while the output has several viable options such as adsorption isotherm, total amount adsorbed, mass flux due to adsorption, heat of adsorption or heat flux. In this instance, the choice was made to focus on mass flux resulting from adsorption. This selection aligns with the hybrid model's approach of calculating the source term for heat flux based on the mass flux data. The distribution of the data points influences the results of the regression analysis and an adequate number of data points for each input variable distributed over the entire range of data points for each variable has to be considered. For the pressure data the logarithmic of the pressure values are used to ensure a better distribution along the different scales of magnitude. All data points are scaled by the maximum value in the range.

A five-fold cross-validation method is used for both teaching and validation phases. The entire process of data generation and machine learning model development is carried out using Octave [36] and Matlab [37], as facilitated by the functionalities available in these software tools.

2.3.2. Teaching and validation

A series of different machine learning techniques are applied to the training and validation data sets. The methods used are polynomial fitting, decision trees, neural networks (NN) and Gaussian process regression (GPR), as shown in Table 2. The best results are for NN and GPR. Thus the analysis is continued with variations of NN and GPR.

For the NN model, a narrow architecture is employed, experimenting with different activation functions including rectified linear unit, sigmoid, and tanh, along with variations in weights and biases. For GPR, a comparison is made among various exponential and Matérn kernel functions. Here the basis function is constant and an isotropic kernel is used. With regard to kernel scale, signal standard deviations and sigma the preset values and algorithms in the Matlab regression learner are used.

The outcomes of employing NN and GPR for machine learning are evaluated side by side. Various models applied in the regression analysis exhibit distinct performance levels in terms of accuracy and computational demands, see Table 3. These differences are systematically documented and compared in Table 3. In order to compare the model accuracy the root mean squared error (RMSE) is used and in order to assess the model performance R-squared (R^2) is used, the values are computed as implemented in Matlab [37].

2.3.3. Comparison GPR and NN: deviations for validation and testing

Utilizing GPR leads to higher computational demands, yet it consistently delivers more accurate outcomes compared to NN, regardless of the specific model parameters applied to both GPR and NN. For NN, optimal performance was observed with a configuration of a single layer and five neurons, employing the rectified linear unit as the activation function and 1000 iterations. Additional adjustments to weights,

Table 2

Comparison of regression with polynomial fitting, trees, neural networks (NN) and Gaussian process (GPR). Root mean square error (RMSE) and coefficient of determination R^2 for validation data sets. Prediction of adsorbed mass hydrogen in AC.

ML method	Fitting	Tree	NN	GPR
RMSE (Validation)	0.003	0.002	0.001	0.0002
R^2 (Validation)	0.999	1.00	1.00	1.00

Table 3

Comparison of regression with narrow neural networks (NN) and Gaussian process (GPR). NN with 1 layer of 5 neurons, GPR with exponential kernel function. Root mean square error (RMSE) and coefficient of determination R^2 for validation and test data sets. Prediction of adsorbed mass hydrogen in AC.

ML method	Neural Network	Gauss Process
RMSE (Validation)	$1006 \cdot 10^{-6}$	$97.3 \cdot 10^{-6}$
R^2 (Validation)	1.00	1.00
Training time (s)	9.79	54.78
Prediction speed (obj/s)	290.000	55.000
RMSE (Testing)	$5020 \cdot 10^{-6}$	$1780 \cdot 10^{-6}$
R^2 (Testing)	1.00	1.00

biases, or an increase in neurons and layers as well as increasing the number of iterations did not markedly improve accuracy. In contrast, the exponential kernel function was identified as the most effective for GPR, balancing accuracy with computational efficiency.

The better RMSE in training but not in testing indicates a potential overfitting issue, especially with regard to the GPR method using the exponential kernel function. For GPR the kernel parameters are preset and optimized by a heuristic approach implemented in Matlab [37]. This procedure is used here without any further input. Although the GPR model seems not optimal due to overfitting, the accuracy as well for the validation as for the testing data is still superior to the other models and thus no further optimization of the GPR model is undertaken. Here the learning-validation and test data are generated as two different data sets and the tendency for overfitting is for all ML models applied to these data sets. A subsequent trial shows that if the learning-and validation data and the test data are generated randomly from the same data set the difference between validation RMSE and testing RMSE decreases. Thus further investigations into the size and structure of the training, validation and test data is advisable.

The best performing NN and GPR models are directly compared in Table 2. In Fig. 3 the predicted values by the NN model are compared to the values of the physical model and in Fig. 4 the predicted values by the GPR-model are compared to the values of the physical model.

For validation purposes, average temperature and pressure values obtained from the ML model are benchmarked against those from a traditional physical model during the process of charging a hydrogen tank. Given that errors can accumulate over the course of a simulation, achieving a mean deviation below 3 %, with R^2 values exceeding 0.995 and RMSE values below 0.002, is mandatory for maintaining fidelity. Models that fail to meet these accuracy benchmarks result in greater deviations.

As the GPR model has the most accurate results, compare Figs. 3 and 4, it is also the computationally most expensive approach. Nevertheless accuracy is paramount due to the accumulating errors during the computation of the charging and discharging cycles. Consequently, the GPR model has been selected for integration into the CFD framework. This GPR-based approach is also adopted for developing ML models for other combinations of adsorbates and adsorbents. Here further investigations into the minimum accuracy needed for these computations would facilitate the decision for the most suitable ML model with regard to the trade off of accuracy and computational costs.

2.4. Hybrid model implementation

The ML-derived equations were integrated into OpenFOAM as source terms, impacting both the mass and energy balance equations. These source terms are crafted to respond dynamically to the local conditions of temperature, pressure, and concentration.

2.5. Comparison

In order to validate the hybrid model the charging and discharging of a 2.8 L tank of activated carbon with hydrogen at ambient conditions is

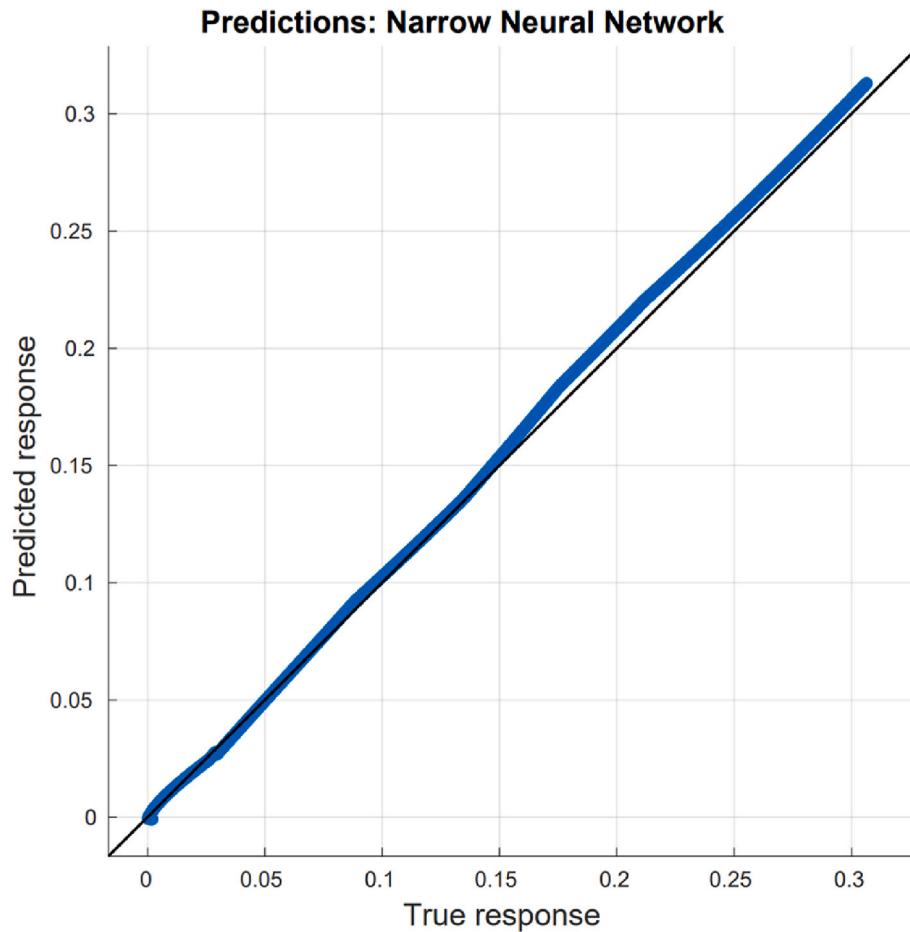


Fig. 3. Predicted values of the best NN-model compared to values of the physical model for the test data set.

simulated. For 442 s the tank is charged with 0.0436 g/s, then the tank rests for 2600 s and finally it is discharged with 0.0219 g/s until 3907 s. These results are compared to experimental data and the results of a simulation of a physical model (CFD with adsorption described by DA isotherm), [14,19].

The computed pressure and temperature in the middle of the tank as a function of time is shown and compared to the results of the physical model and the experimental data from the literature, see Figs. 5 and 6. The qualitative and the quantitative agreement is good and has a similar order of magnitude as the measurement errors. Thus the hybrid model can accurately predict hydrogen adsorption in activated carbons.

In addition the deviations of the hybrid model to the measured data is compared to the deviations of the physical model to the measured data in Figs. 7 and 8. The deviations for both approaches are of similar order of magnitude. Thus the quality of the results are similar regardless of the model used. The computational effort for the data driven model is significantly lower, the computational time needed by the physical adsorption model is approximately two time higher. Thus the data driven model yields advantages when many computations have to be performed, as for optimizations.

Here the most accurate model, i.e. GPR model, is implemented. The implementation of a less accurate model is computational even less expensive. Nevertheless, with these models even small errors accumulate leading to significant deviations in the long run of a charging and discharging process.

3. Results

Using the hybrid model, the dynamics of hydrogen charging and

discharging in activated carbon tanks under various configurations and hydrogen flow rates are analysed, as depicted in Figs. 9 and 10. The effects of tank porosity, geometry, flow channels, and strategies for enhancing heat transfer can be analysed. Densely packed adsorbent materials with low porosity exhibit a pressure gradient across the material. This gradient leads to a corresponding temperature gradient and a heterogeneous distribution of adsorbed hydrogen, as illustrated in Fig. 9. Improvements in heat transfer, both at the tank walls and internally, significantly affect the temperature and concentration distributions within the tank, as shown in Fig. 10. Flow channels designed to direct the hydrogen gas flow as well as devices for thermal management impact the distribution within the tank.

The charging and discharging of a 33 L tank of AX21 at ambient conditions is analysed. Bundles of such tanks may be used in vehicles as fuel tanks. At ambient temperatures the amount of hydrogen stored does not meet the DOE targets.

For the activated carbons considered in this study the temperature and pressure variations influence the amount of adsorbed hydrogen only slightly, the main influence is on the hydrogen which is compressed in the voids of the porous medium.

In the case of fast charging the tank heats up leading to an increase in the temperature and the compressed gas density. Thus the tank storage capacity is only partially used with fast charging. Here additional cooling is required. If the tank operates at ambient temperatures appropriate measures may be taken to increase the heat transfer at the tank walls: With forced convection by ventilating the tank surface the heat transfer coefficient increases. An increase of the exterior surface of the tank is achieved by using fins. An energy efficient alternative for vehicle tanks is overnight charging at low flow rates.

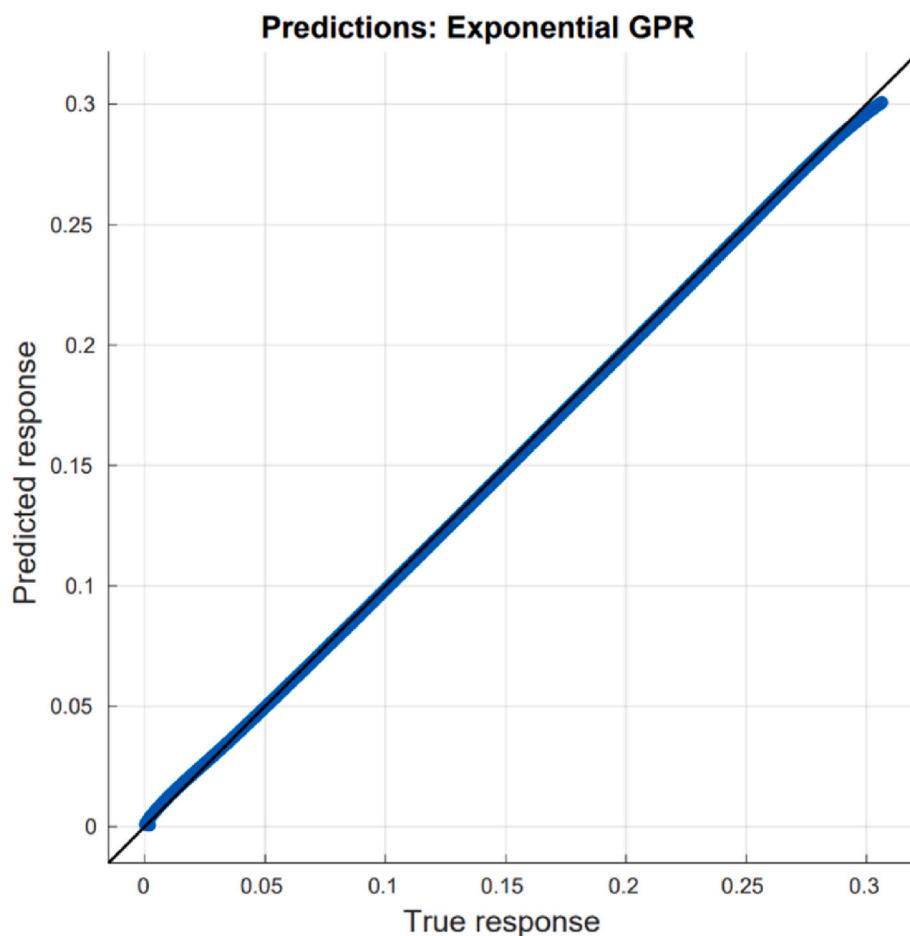


Fig. 4. Predicted values of the best GPR-model compared to values of the physical model for the test data set.

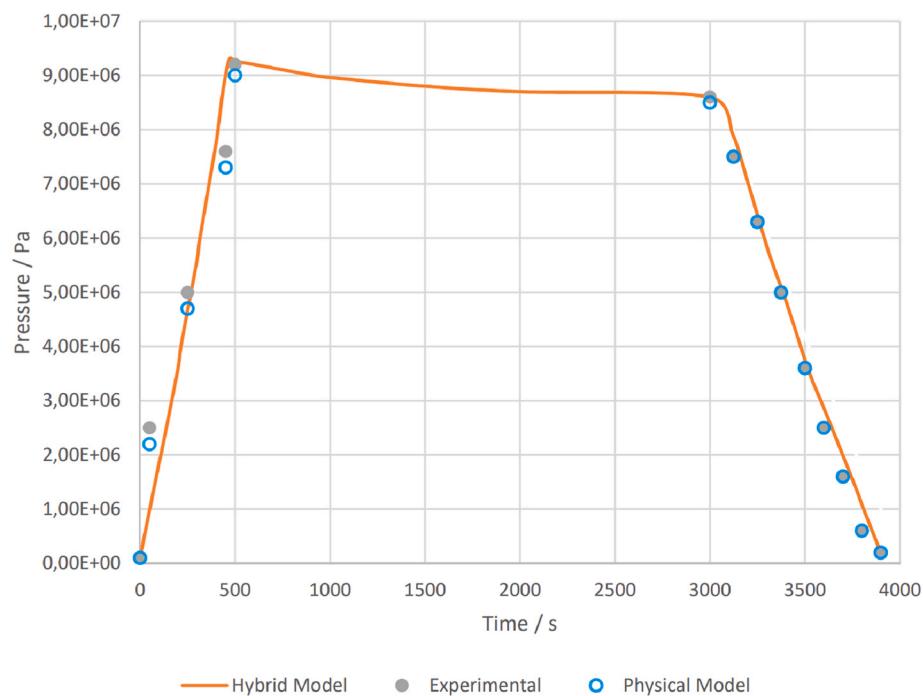


Fig. 5. Pressure distribution for charging and discharging of a 2.8 L adsorption tank with hydrogen. Reference data from Ref. [14].

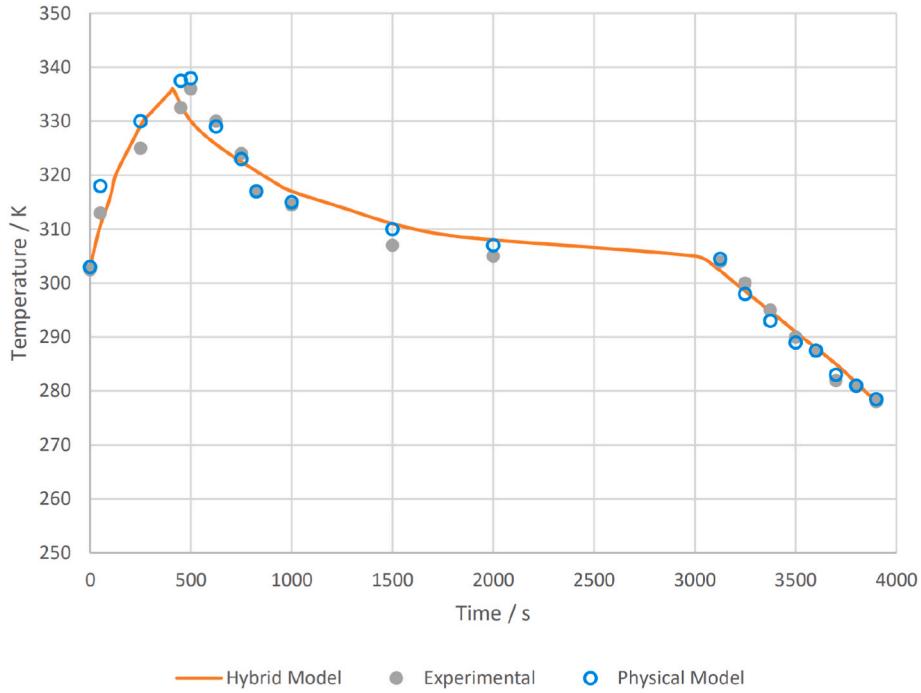


Fig. 6. Temperature distribution for charging and discharging of a 2.8 L adsorption tank with hydrogen. Reference data from Ref. [14].

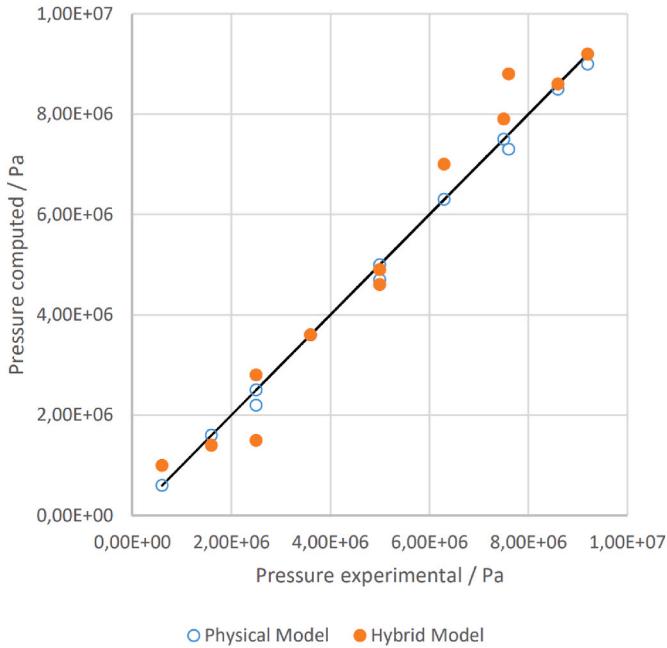


Fig. 7. Comparison of measured and computed pressure values during the charging and discharging of a 2.8 L adsorption-tank with hydrogen. The values are computed with a physical model and a hybrid model incorporating a ML model for the adsorption. Reference data from Ref. [14].

Similarly in the case of fast discharging the tank cools down and the compressed gas density decreases and the exit pressure has to be further decreased to maintain the desired mass outflow. Some additional hydrogen still rests in the tank.

The influence of power charging of a 33 L tank with AX-21 at ambient conditions is shown in Fig. 11, the influence of the discharging rate for this tank is shown in Fig. 12. For high mass flow rates, i.e. bigger than 2 g/s, additional cooling should be applied. With forced convection a mass flux of 8 g/s is possible without relevant penalties.

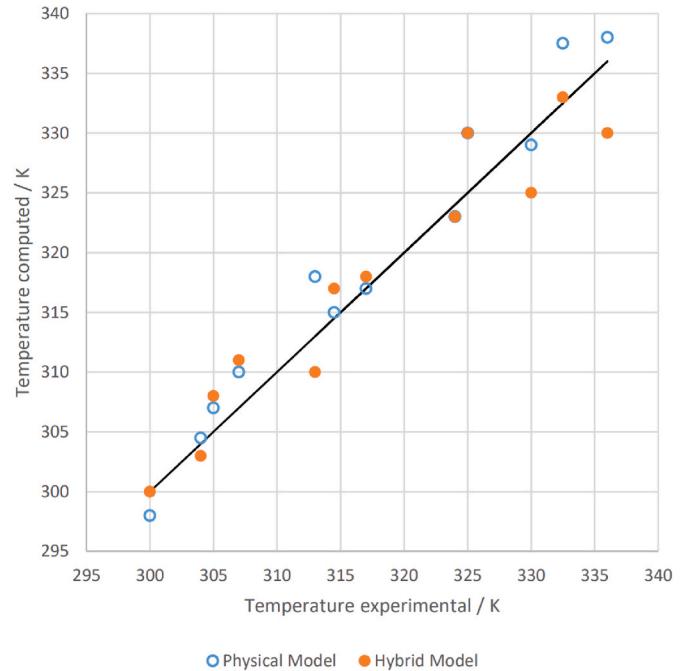


Fig. 8. Comparison of measured and computed temperature values during the charging and discharging of a 2.8 L adsorption-tank with hydrogen. The values are computed with a physical model and a hybrid model incorporating a ML model for the adsorption. Reference data from Ref. [14].

Trying to balance the temperature distribution in the interior is of lesser significance.

The pressure gradient is of importance at the low pressure levels. The overall performance of the tank is influenced for fast discharging as some hydrogen still rests in the tank.

Thus the operating conditions of adsorption tanks would be similar to those of the compressed hydrogen storage tanks already in use, the difference would be the greater amount of hydrogen stored at somewhat

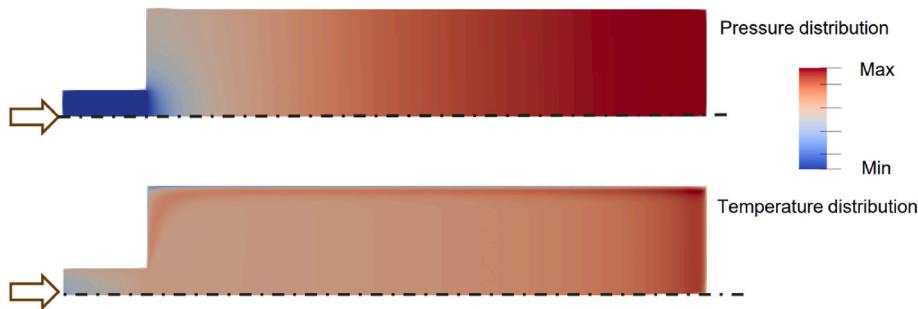


Fig. 9. Charging of a 196 L tank with activated carbon with 20.4 L per second. A pressure gradient develops due to the very low porosity of the activated carbon monolith. The pressure gradient persists with increasing pressure in the tank.

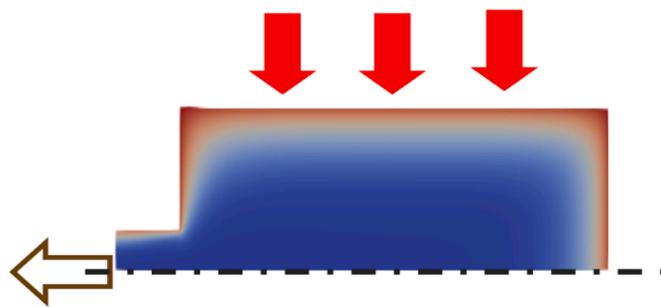


Fig. 10. Discharging of a 33 L tank with activated carbon with 0.3 g/s. Temperature distribution in the tank. The heat transfer at the tank walls is enhanced by forced convection, leading to a radial temperature gradient in the near wall region of the tank.

lower pressures in the adsorption tanks.

In order to achieve the DOE targets, an adsorption tank with AX81 would have to be cooled to under 100 K. The influence of the system temperature on the storage capacity of a 33 L tank of AX21 is shown in Fig. 13. The resulting heat of adsorption has to be diverted, resulting in the necessity of a thermal management system in addition to the cooling requirements

A less elaborate solution is only possible using adsorption material with a storage capacity superior to AX21 (i.e. the micropore volume of

this carbon would have to be three times higher). During fast charging and discharging temperature differences up to 80 K occur in the tank thus requiring appliances for heat management.

The hybrid CFD model can easily compute the spatial distribution of temperature, concentration and pressure, which are more significant for cryogenic applications or hydrogen storage in hydrides with higher heats of sorption or materials with very high storage capacity conforming or exceeding the DOE targets. For the design and optimization of heat management appliances in tank systems an efficient and effective CFD tool, as presented here, is a prerequisite.

These insights offer valuable guidance for optimizing tank design. Moreover, they enable the evaluation of the possibilities and limitations in lumped parameter modelling, which might be considered as a fast alternative modelling approach. Extensive datasets generated via the hybrid model facilitate the creation of digital twins for the adsorption tanks, serving as a powerful tool for further exploration and optimization.

4. Conclusion

4.1. Summary

ML-models are developed to simulate the adsorption of hydrogen in activated carbon, exploring various strategies for model creation based on dataset size and structure, as well as instructional methods. The investigation covers different ML regression algorithms and the

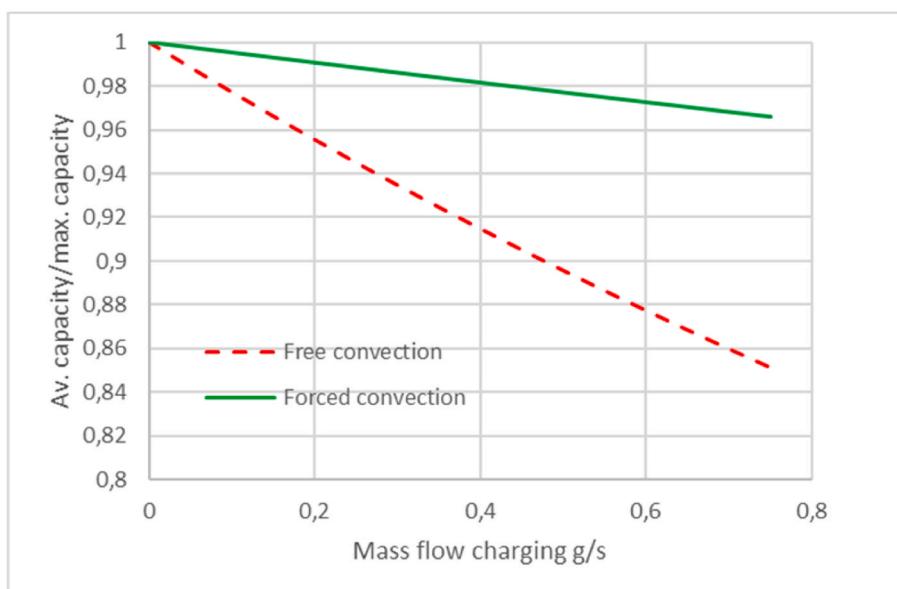


Fig. 11. Charging of a 33 L tank with AX-21. With increasing mass flow rate the storage capacity is only partially used.

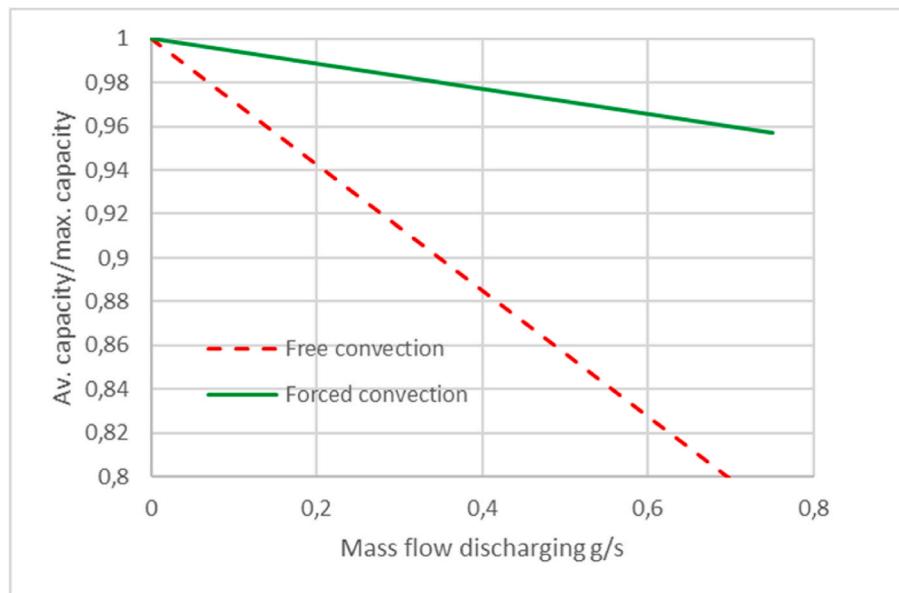


Fig. 12. Discharging of a 33 L tank with AX-21. With increasing mass flow rate the stored hydrogen is only partially available.

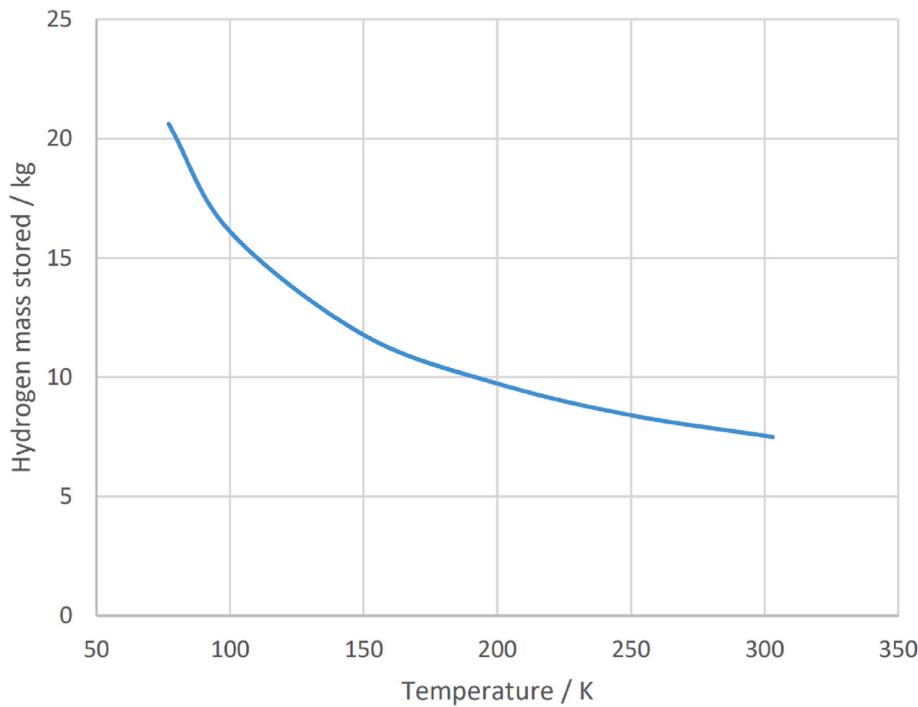


Fig. 13. Mass of hydrogen stored at 350 bar in a 33 L type IV tank filled with AX21. Influence of system temperature.

structuring of input and output data. Given the limited size of the datasets, the GPR method emerges as the most effective in terms of accuracy and precision. Utilizing GPR with appropriately scaled variables proves highly advantageous for the regression model, where high accuracy and precision are crucial to prevent error accumulation during computation. The datasets employed are derived from a model based on the Dubinin-Astakhov isotherm; however, this methodology also allows for the incorporation of data from other modelled adsorption isotherms and also from measured adsorption isotherms. These models are integrated into a CFD code to assess the distribution of concentration, temperature, and pressure during the charging and discharging of activated carbon tanks with hydrogen, aiming to optimize tank design and

operating conditions. For higher storage capacities, as targeted by the DOE, and in rapid fuelling and defueling scenarios, the heat of adsorption becomes significant, necessitating effective thermal management of the hydrogen tank.

As a first step the ML-models derived here are for specific materials (hydrogen and activated carbon). Its accuracy is comparable to physical models. It is applicable in the data range, i.e. operating conditions and materials, used for teaching and validation. Using this approach ML models for different materials may be derived.

As a result of this novel modelling approach the following issues arise, which are to be assessed in future investigations:

- **Size and structure of teaching, validation and testing data sets**
- Accuracy needed for the adsorption model implemented in the CFD code to assess trade off accuracy – computational cost
- Investigation and optimization of appropriate NN model parameters
- Investigation and optimization of appropriate GPR model parameters

4.2. Outlook

The approach shown here can easily be applied to the storage of hydrogen in other adsorbents, i.e. metal hydrides and other adsorbates i.e. methane. As the approach is data driven, it is also feasible to model chemisorption, i.e. metal hydrides, in a similar way.

Extending this data driven approach, it is possible to compute the sorption of gas mixtures, which is of importance in several applications, by extending this data driven approach by a variable describing the composition. As a physical model sorption of mixtures are computationally more challenging, due to the additional iterations needed to determine the equilibrium. As a further step, parameters describing the adsorption (e.g. micropore volume, affinity coefficient, adsorption energy) may be incorporated in the ML model as input variables, leading to a generalized adsorption ML model, thus facilitating the analysis and development of novel sorbent materials.

The hybrid model that employs an ML-based description of the adsorption process coupled with a CFD model is set for further exploration and adaptation to additional applications. This methodology facilitates the easy generation of data under various operating conditions and can be expanded to other sorbents, gases, gas mixtures, and chemisorption processes. The next phase will involve incorporating adsorption and material properties as variables within the ML model, with subsequent exploration into ML models for chemical bonding of hydrogen and other gases in hydrides. Efforts to refine the generation of ML models and their integration into hybrid models will continue, focusing on the variables' formulation and ML methods to enhance model accuracy and efficiency with a minimal amount of data required. Further validation through experimental data and its use as input for ML models will be pursued. This approach also promises to simplify the generation of extensive datasets for hydrogen adsorption in tanks, thereby facilitating numerical optimization and the potential development of a digital twin based on computational results. In the future this approach may also be used for other industrial processes involving adsorption, like carbon capture, thermal energy storage or upgrading of methane.

CRediT authorship contribution statement

Georg Klepp: Visualization, Validation, Methodology, Investigation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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