# Hydrogen Adsorption Model Using Neuro-fuzzy System

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*Abstract*
A study of hydrogen gas adsorption onto carbon based material is essential in hydrogen storage research for future on board vehicles. Presently many experiments have been conducted to find out hydrogen adsorption capacity and several models of hydrogen adsorption have been work out to fit in with the experimental data. However, the current models developed were based on assumptions made for examples adsorption at supercritical conditions, types of carbon surface and structures as well as interaction of $H_2$ molecules and adsorbent molecules. This paper highlights the modelling of the hydrogen adsorption isotherm onto activated carbon using an assumption-free model which is neuro-fuzzy system. The hydrogen adsorption isotherm model was developed using equilibrium data at temperature range from 173 K to 253 K and pressure range from 0.16 to 6.1 MPa. The average error difference between the equilibrium data and predicted model is found to be less than 3%. The model is validated at 193 K and 233 K. Thus, the developed model is applicable to predict the adsorption isotherms of hydrogen at the temperature range from 173 K to 253 K and pressure range from 0.16 to 6.1 MPa. It is comparable with the established models used to develop adsorption isotherm.

Key-words: assumption-free model, hydrogen adsorption model, neuro-fuzzy system

## 1 Introduction

Hydrogen gas ($H_2$) has been known as potential alternative energy to replace oil and gas for on board vehicles. This is because $H_2$ gives pollution-free since its combustion will result in steam i.e. water vapour. In addition, $H_2$ possess high-energy content compared to its weight. However, the energy content compared to its volume is rather low. Thus, one of the critical issues that need to overcome in hydrogen-based transportation system is $H_2$ storage. Such problem provides a great challenge as compared to storage of gasoline, which is liquid.

$H_2$ can be stored in several different ways such as compression, liquefaction, or using storage medium like carbon based materials, glass microspheres, metal hydrides or gas clathrate hydrates. As for transportation, carbon based material has been identified as promising adsorbent to store $H_2$. This is due to its light-weight and high surface area, which provides low volume in $H_2$ storage.

Carbon based material could be in the form of activated carbon, super activated carbon, carbon nanotubes, graphitic nanofibres, carbon monolith, etc. All these materials have been synthesized and used in investigating $H_2$ adsorption for storage purposes. Apart from experiment, many researchers have come out different types of model in describing the behaviour of gas

adsorption in microporous adsorbent. Their predictions are based on assumptions made due to several reasons such as adsorption at supercritical conditions, types of carbon surface and structures as well as interaction of $H_2$ molecules and adsorbent molecules.

As a result, hydrogen adsorption can be predicted based on model developed such as Grand Canonical Monte Carlo (GCMC) [4], Ono-Kondo fit model [2,6], Linear Comprehensive Supercritical Adsorption Model [9], Dubinin-Astakhov equation [10] and Langmuir Model [11] (refer to Table 1). However, each model has its own limitations due to several assumptions made. Hence, a new approached in adsorption model is introduced to improve and simplify the predicted model.

Unlike with any of the conventional models in Table 1, an assumption-free model of hydrogen adsorption can be developed using Neuro-fuzzy system. Such programming has been used before in predicting methane adsorption onto activated carbon [4,7].

Table 1: Comparison of hydrogen adsorption using different models

| Operating Conditions | Adsorption Capacity | Model Applied | References |
|---|---|---|---|
| 293.15 K, 2.66 MPa | 4.6 kg$H_2$ /m system | Grand Canonical Monte Carlo | Chong Gu et. al (2001) |
| 77 - 298 K, 19 kg/m$^3$ | 53 g/kg | Ono-Kondo Fit Model | Bénard et. al. (2001) |
| 93 – 293 K, 7 MPa | 54 mmol/g | Linear Comprehensive Supercritical Adsorption Model | Zhan et al. (2002) |
| 93 – 293 K, 6.1 MPa | 49 mmol/g | Dubinin-Astakhov equation | Zhan et. al (2004) |
| 93 – 273 K, 6 MPa | 5.3 wt% | Ono-Kondo Fit Model | Poirier et. al. (2004) |
| 298.15 K | 3 mass% | Langmuir Model | Züttel et. al. (2004) |

The objective of the study is to predict hydrogen adsorption isotherm onto activated carbon at temperature range from 173 K to 253 K and pressure from 0.16 MPa to 6.10 MPa with adsorption capacity at a maximum of 35 mmol/g using neuro-fuzzy system.

## 2 Theory

Neuro-fuzzy is the combination of explicit knowledge representation of fuzzy logic with the learning power of neural network. Neural network is good at recognizing patterns but not good at explaining how they reach their decisions. On the other hand, fuzzy logic systems is good at explaining their decisions but they are not automatically acquire the rules that use to make those decisions. Hence, as a result of this combination, neural network can learn from data sets while fuzzy logic solutions are easy to verify and optimize [1].

In this study, the simulation of hydrogen adsorption model was done using fuzzyTECH 5.52 Professional Edition that has a NeuroFuzzy module. The neuro-fuzzy system applies the technology of neural network to produce fuzzy logic rules and membership functions automatically [3]. It contains FuzzyCluster module that is suited for preparing training data for the NeuroFuzzy module.

To create a complete fuzzy logic system, Fuzzy Design Wizard (FDW) is set up. FDW allows the user to define variables i.e. FDW can extract the number and the name of the variables as well as the data range of each variable from the appropriate sample data file [7]. In addition, FDW enables the user to define defuzzification and rule blocks. Defuzzification is a transformation from a fuzzy set to a crisp number. In this

modeling, Centre of Maximum (CoM) is chosen since neuro-fuzzy training uses error gradients to determine a direction of optimization [7].

## 3 Methodology

There were several steps in developing model using neuro-fuzzy system. The first step was to obtain the training data. In this case, the experimental data i.e. hydrogen adsorption isotherm data was obtained from the literature (Zhan, 2004) at temperature range from 173 K to 253 K. The data was identified as TISO system that consisted of two input variables: temperature (in K) and pressure (in MPa) and one output variable: adsorption capacity (in mmol/g).

The neuro-fuzzy system training process started with initial fuzzy logic system. Hence, the second step was to create fuzzy logic system by using a Fuzzy Design Wizard (FDW), which started a sequence of dialogs that would guide the user to create a complete fuzzy logic prototype. The advantage of coming up with such initial rules was that it enhanced the performance of the learning process. For example IF temperature=high AND pressure=low THEN capacity=low. However, in this case the FDW analysed the data sets and proposed a system structure. Once the structure was accepted, FDW created a system with default membership function definitions and default rules.

The third step was to optimise the performance by parameterized the neuro-fuzzy learning method. There were two important parameters. One was the learn rate for rules and the other one was the learn rate for membership functions. These parameters defined how much the Neuro-fuzzy algorithm modified a rule or a membership function in each learning step.

Training phase or simulation took place in step 4 in which the Neuro-fuzzy system modified rules and membership functions in order to obtain a predicted result as close to the experimental data as possible.

In step 5, after the simulation had been performed, the predicted training data ($Q_{i,predicted}$) obtained was compared with the experimental data ($Q_{i,experimental}$) by calculating the mean absolute relative errors (*MAREs*). This was done in order to ensure reliability and accuracy of forecast data that produced by the system. If the *MAREs* were less than 5%, the forecasted data was displayed as the result.

$$MAREs\ (\%) = \left(\frac{100}{N}\right)\sum_{i=1}^{N}\left|\frac{Q_{i,experimental} - Q_{i,predicted}}{Q_{i,experimental}}\right| \quad (1)$$

For *MAREs* more than 5%, step 6 that was optimisation and verification was taken place. The results of neuro-fuzzy system could be manually optimised by tuning the membership functions. After modification, the training was started all over again with the same sample data sets. A complete flow of neuro-fuzzy approach is shown in Figure 1.

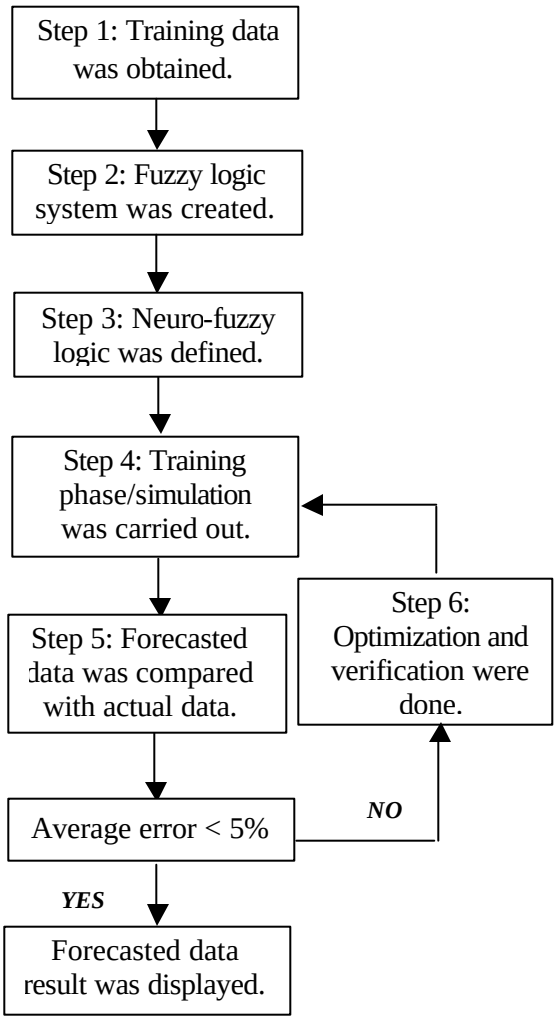


Figure 1: Flowchart of system design approach

To verify the neuro-fuzzy capability as assumption-free model in predicting the hydrogen adsorption isotherm, the model was validated between temperature 173 K and 273 K. The temperature of 193 K and 233 K were chosen since the equilibrium data was available in the literature (Zhan, 2004).

## 4 Results and Discussion

The prediction was done specifically for isotherm data of hydrogen adsorbed onto activated carbon. It was done at three different temperatures that are 173 K, 213 K and 253 K. The pressure range was from 0.16 MPa up to reference pressure at 6.10 MPa. A minimum of fourteen (14) points of adsorption data of each temperature was taken from literature in order to generate the isotherm model

The model was reviewed in order to achieve targeted relative error of <5%. The final limit of range for all parameters contributed to the prediction of hydrogen adsorption isotherm onto activated carbon were stated below:

For temperature (Input 1), the membership function was divided into three terms that were:

i. Low : 173.0 – 287.6 K
ii. Medium : 182.9 – 253.0 K
iii. High : 236.4 – 253.0 K

For pressure (Input 2), the membership function was divided into three terms that were:

i. Low : 0.16 – 5.44 MPa
ii. Medium : 0.13 – 5.55 MPa
iii. High : 0.53 – 6.10 MPa

For amount adsorbed (Output), the membership function also was divided into five terms that were:

i. Very Low : 0 – 3.91 mmol/g
ii. Low : 1.45 – 16.78 mmol/g
iii. Medium : 11.67 – 30.37 mmol/g
iv. High : 20.00 – 34.13 mmol/g
v. Very High: 23.24 –35.00 mmol/g

After the system of the adsorption isotherm model was developed, both forecasted data and actual experimental data were compared between each other in order to test the reliability and accuracy of the system (see Figure 2).

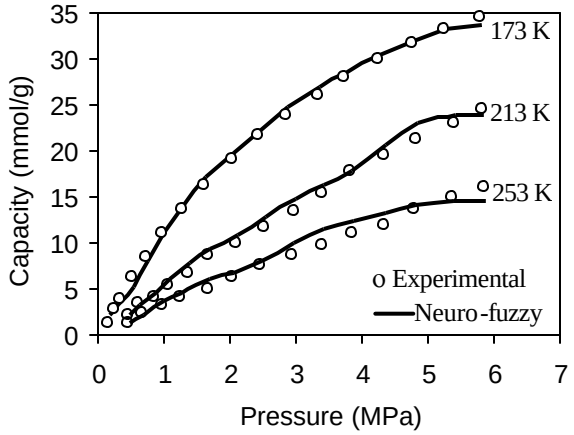


Figure 2: Comparison between isotherm model of hydrogen adsorption using Neuro-Fuzzy with experimental data (Zhan, 2004).

The model developed shows similar trend with the equilibrium data (Zhan, 2004) and it can be used to predict the hydrogen adsorption isotherm at the temperature range of 173 and 253 K and pressure range of 0.16 to 6.1 MPa with adsorption capacity up to 35 mmol/g. To support the results, an error analysis was done in order to ensure the accuracy and the reliability of the model developed.

Table 2 shows the calculated average error for the respective temperature. It can be seen that for each temperature, the relative error is less than 3%. It shows that the assumption-free model by neuro-fuzzy system is applicable for developing adsorption isotherm.

Table 2: *MAREs* results obtained from Neuro-Fuzzy System.

| Isotherm Model Temperature (K) | *MAREs* (%) |
|---|---|
| 173 | 0.76 |
| 213 | 2.75 |
| 253 | 2.66 |

Figure 3 shows the comparison between the neuro-fuzzy model and the equilibrium data of hydrogen adsorption isotherm at 193 K and 233 K.

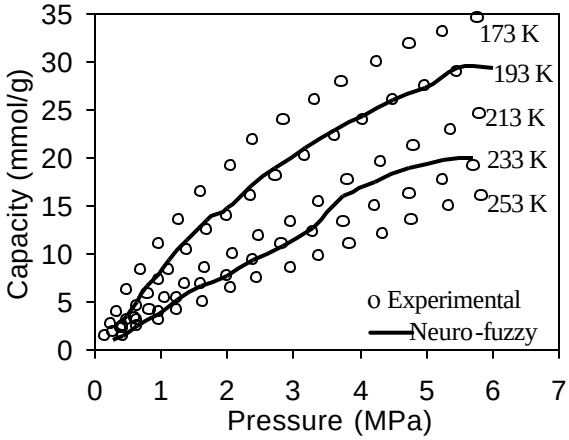


Figure 3: Model validation of hydrogen adsorption isotherm at temperature of 3193 K and 233 K.

In addition, from the model developed, it is possible to predict the adsorption isotherm at other temperature within the temperature range of 173 and 253 K and pressure range of 0.16 to 6.1 MPa as shown in Figure 4.

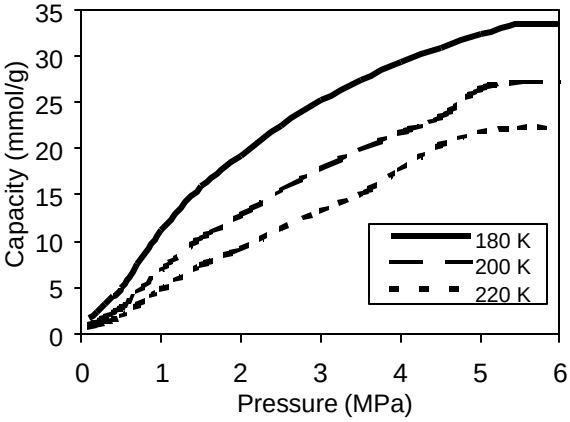


Figure 4: Prediction of hydrogen adsorption isotherm at various temperatures.

## 5 Conclusion

The prediction of adsorption isotherm for hydrogen gas onto activated carbon was successfully implemented by using neuro-fuzzy system with an average error of less than 3%. The isotherm predicted using neuro-fuzzy approach and experimental data show almost similar trend with each other. It can be concluded that the assumption-free model using neuro-fuzzy system is able to predict the adsorption isotherm of methane gas onto activated carbon within temperature range from 173 to 253 K and pressure range from 0.16 to 6.10 MPa.

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