

An Artificial Neural Network Based Model for Predicting H₂ Production Rates in a Sucrose-Based Bioreactor System

Nikhil, Bestamin Özkaya, Ari Visa, Chiu-Yue Lin, Jaakko A. Puhakka, and Olli Yli-Harja

Abstract—The performance of a sucrose-based H₂ production in a completely stirred tank reactor (CSTR) was modeled by neural network back-propagation (BP) algorithm. The H₂ production was monitored over a period of 450 days at 35±1 °C. The proposed model predicts H₂ production rates based on hydraulic retention time (HRT), recycle ratio, sucrose concentration and degradation, biomass concentrations, pH, alkalinity, oxidation-reduction potential (ORP), acids and alcohols concentrations. Artificial neural networks (ANNs) have an ability to capture non-linear information very efficiently. In this study, a predictive controller was proposed for management and operation of large scale H₂-fermenting systems. The relevant control strategies can be activated by this method. BP based ANNs modeling results was very successful and an excellent match was obtained between the measured and the predicted rates. The efficient H₂ production and system control can be provided by predictive control method combined with the robust BP based ANN modeling tool.

Keywords—Back-propagation, biohydrogen, bioprocess modeling, neural networks.

I. INTRODUCTION

THE fossil fuel dependency of energy economy today results in global warming, air pollution and environmental and health problems. Hydrogen (H₂) produced from renewable energy sources offers a clean alternative for the fossil fuels [1]. Besides uses as an energy carrier, H₂ has multiple uses in industrial applications, such as production of lower molecular weight compounds, saturation of compounds, cracking of hydrocarbons or removal of sulfur and nitrogen compounds, O₂ scavenger to prevent corrosion and oxidation, and coolant

in electrical generators [2]. Today, several techniques for sustainable H₂-production exist including microbiological fermentation processes [3].

Microbiological dark fermentation, involving mixed microbial cultures, can be used to produce H₂ from biomass or organic waste materials [4], [5]. H₂ production is an intermediate step in the anaerobic degradation of organic material. H₂ is produced in order to maintain the electron balance in the anaerobic system [6]. The gases (H₂ and CO₂) and organic acids such as acetate, butyrate, propionate and valerate, and alcohols (e.g. ethanol) are the end products of the bioprocess [6]-[8]. In this study, input and monitoring parameters and end-products (organic acids, alcohols) and biomass concentrations have been used to predict H₂ production rate.

It is very important to predict H₂ production rates by a comprehensive model for the design, monitoring and management of biohydrogen producing bioreactors. ANN models may be successfully applied in biohydrogen production systems and are very effective in capturing the nonlinear relationships existing between variables (multi-input/output) in complex system like biohydrogen production. This study aims at using ANN capabilities to predict H₂ production rates in completely stirred tank reactor (CSTR) and thereby managing the bioreactor. The proposed ANN based-model predicts H₂ production rate from hydraulic retention time (HRT), recycle ratio, sucrose concentration, sucrose degradation, biomass concentrations, pH, alkalinity, oxidation-reduction potential (ORP), ethanol, acetate, propionate and butyrate concentrations.

Recently, numerous studies have reported the use of ANN models in environmental engineering applications [9]-[14] which were successively applied and excellent match obtained by this robust tool, yet most of these models are applicable to only output data prediction. In addition, there are several works for controlling of complex bioprocess and biosystems in environmental and industrial applications [15]-[17]. For this work, it was hypothesized that a method can be developed that can propose the predictive control method of H₂ production rate by combining BP based ANN model predictions. Thus the purpose of this study was to evaluate and develop a combined BP based control predictive method for H₂-fermenting systems.

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II. EXPERIMENTAL

A. Completely Stirred Tank Reactor (CSTR)

The CSTR placed in a water-bath tank (Fig. 1), with a working volume of 4.0 L, was operated for 15 months at $35 \pm 1^\circ\text{C}$. The seed sludge was obtained from Li-Ming Municipal Sewage Treatment Plant (Taichung, Taiwan). The collected sludge was screened with a No. 8 mesh (diam. 2.35 mm) and was preheated at 100°C for 45 minutes to inhibit methanogen or other microorganisms' bioactivity. The seed sludge was acclimated with sucrose at a concentration of 20 g COD/L in a growth medium consisting of [18] (mg/L): NH_4HCO_3 5240, K_2HPO_4 125, $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ 100, $\text{MnSO}_4 \cdot 6\text{H}_2\text{O}$ 15, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ 25, $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ 5, $\text{CoCl}_2 \cdot 5\text{H}_2\text{O}$ 0.125, NaHCO_3 6720. The substrate was stored at 4°C .

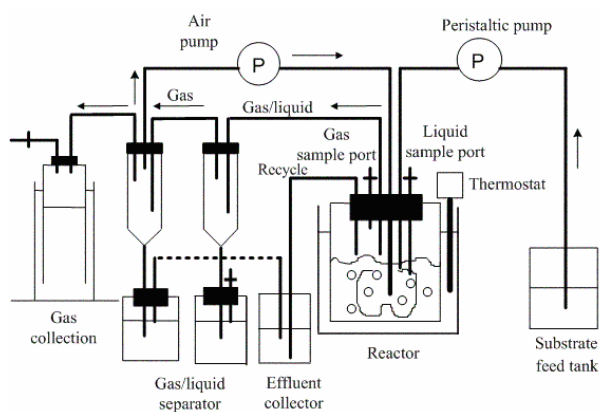


Fig. 1 Configuration of an anaerobic completely stirred tank reactor (CSTR) for continuous H_2 production

Initial CSTR operation was in a continuous feeding mode and hydraulic retention time (HRT) was 12 h. The CSTR pH was controlled and maintained at around 6.7 which was found to be favorable for hydrogen production [19], [20]. When a steady-state condition was reached and the desired data were obtained the recycle ratio or substrate concentration or HRT was reduced. At each run, the CSTR was operated for more than ten times of the HRT to develop a steady-state condition. Steady-state conditions reached when the product concentrations such hydrogen gas content, biogas volume and metabolite concentrations were stable (less than 10 % variation). For each steady-state data measurement, 6-10 samples were analyzed.

TABLE I
THE SUMMARY OF THE REACTOR OPERATIONS

Reactor operation	HRT (h)	Sucrose concentration (g COD/L)	Recycle ratio
Stage I	12	20	0, 0.2, 0.4, 0.6, 0.8, 1
Stage II	12, 8, 6, 4, 2	20	0.2
Stage III	12	20, 25, 32, 40	0.2

Table I shows the experimental stages of this study. The CSTR was routinely monitored for pH, alkalinity, oxidation-

reduction potential (ORP), gas production and composition, sucrose concentration, ethanol concentration, volatile fatty acid (VFA) distribution and volatile suspended solids (VSS) concentrations. The gas volumes were corrected to a standard temperature (0°C) and pressure (760mmHg).

B. Bioprocess Monitoring Analyses

The mixed liquors sampled were centrifuged (900 g, 15 min) and the supernatants were taken for metabolite analysis. VFA and ethanol were analyzed with a gas chromatograph having a flame ionization detector (Shimadze GC-14, Japan). Biogas volume was determined by a gas meter (Ritter, Germany). Biogas composition except hydrogen sulfide was analyzed with a gas chromatograph having and a thermal conductivity detector (China Chromatograph 8700T, Taiwan). Hydrogen sulfide gas was analyzed with a gas chromatograph having a flame photometric detector (capillary column, 150°C ; injection temperature, 150°C ; carrier gas, N_2). Other analytical details for the VFA, ethanol and biogas assays were the same as those in our previous study [21], [22]. Anthrone-sulfuric acid method was used to measure sucrose [23]. The ORP value was measured using a pH/ORP Controller with a silver chloride electrode (Suntex, Taiwan). Other water quality parameters were measured according the procedures of Standard Methods [24].

III. ANN BASED PREDICTION OF H_2 PRODUCTION AND ITS PREDICTIVE CONTROL

Neural networks are composed of simple elements operating in parallel. As in nature, the network function is determined largely by the connections between elements. Commonly neural networks (NN) are adjusted, or trained, so that a particular input leads to a specific target output. In the basic neural networks architecture, the network is adjusted, based on a comparison of the output and the target, until the network output matches the target [25]. Typically many such input/target pairs are used to train a network. Back-propagation (BP) algorithms use input vectors and corresponding target vectors to train an NN. The neural networks with a sigmoid and linear output layer are capable of approximating any function with a finite number of discontinuities [26]. The standard BP algorithm is a gradient descent algorithm, in which the network weights are changed along the negative of the gradient of the performance function [27], [28]. There are a number of variations of the basic back-propagation algorithm, which are based on other optimization techniques, such as conjugate gradient and Newton methods. For properly trained back-propagation networks, a new input leads to an output similar to the correct output. This NN property enables the training of a network on a representative set of input/target pairs and getting well predicting results.

In this case, H_2 -production rate is the most important control parameter and selected as output parameter in CSTR system because of energy recovery and process performance. The main difficulties in control of biological process are the variability of the kinetic parameters, especially in anaerobic

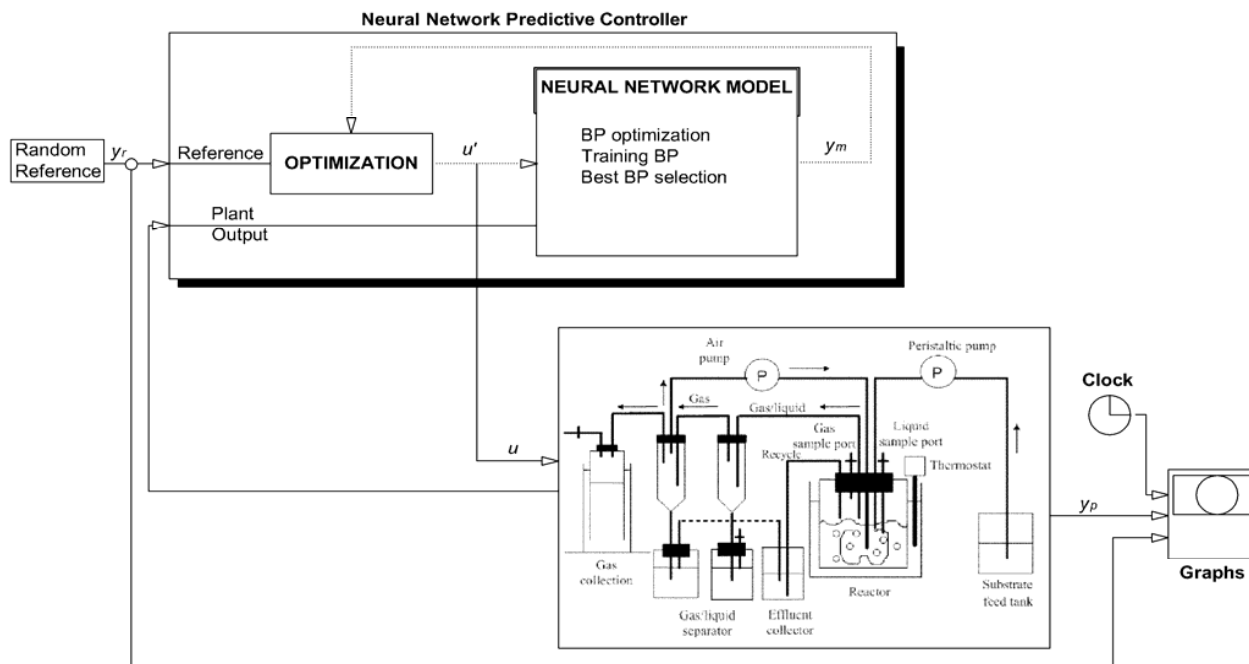


Fig. 2 Neural network predictive control of completely stirred tank reactor (CSTR) for continuous H_2 production (adapted from Matlab)

fermentation system bioprocesses consisting of several steps. Further, one step may be limiting factor for the other steps, such as time-varying influent wastewater characteristics and non-linearity. Hence adaptive and non-linear controller is the excellent choice for biological process control. Due to their impressive capability in dealing with severe non-linearity and uncertainty of a system, the application of neural network method for the design of controllers is promising. In the present study, twelve input variables were used for robust prediction of H_2 production. This study is aimed at obtaining a proper H_2 production control in the complex biological system by proposing a method based on BP algorithms with the NN and model predictive method (Fig. 2). This method is based on the receding horizon technique. The neural network model

$$J = \sum_{j=N_1}^{N_2} (y_r(t+j) - y_m(t+j))^2 + \rho \sum_{j=1}^{N_u} (u'(t+j-1) - u'(t+j-2))$$

predicts the plant response over a specified time horizon; where N_1 , N_2 and N_u define the horizons over which the tracking error and the control increments are evaluated. The u' variable is the tentative control signal, y_r is desired response and y_m is the network model response. The ρ value determines the contribution that the sum of squares of the control increments has on the performance index (Matlab 7 Toolbox).

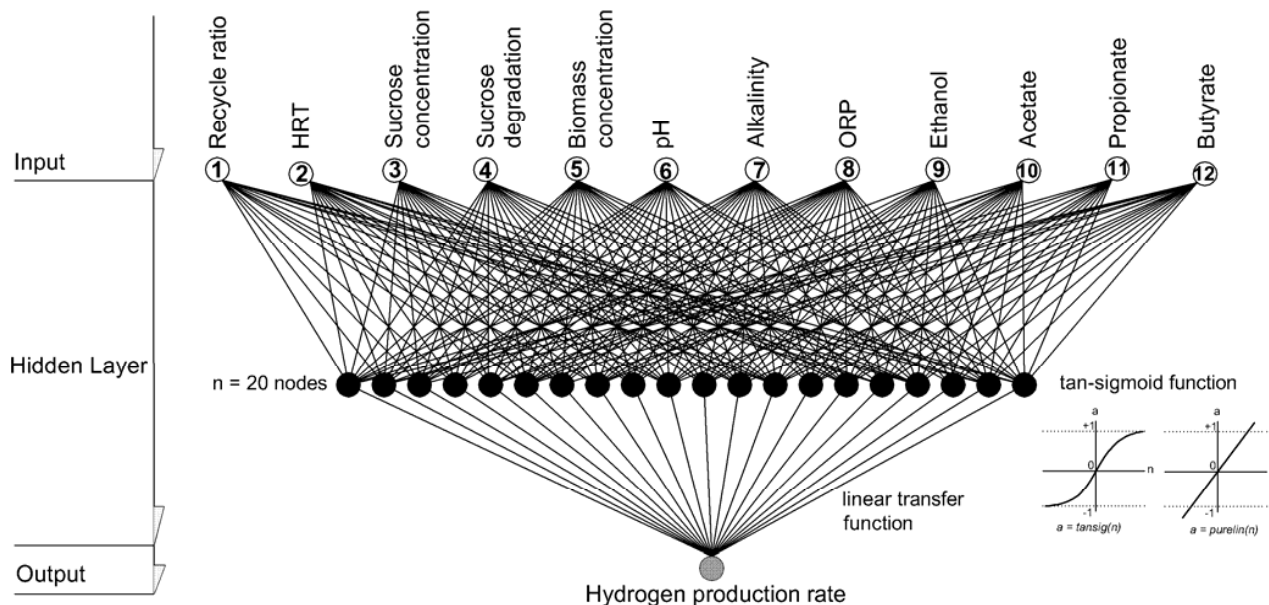
The diagram in Fig. 2 shows the model predictive control process. The controller consists of the NN plant model and the optimization block. The optimization block determines the value of u' that minimize J , and then the optimal u is input to the plant. This method consist of three steps including optimization, BP based ANN model and reactor system. To optimize the H_2 production rate with this method, CSTR system can be operated with the optimum operational

conditions (inputs) such as recycle ratio, HRT, pH, alkalinity, and sucrose concentration. In this system, if the H_2 production is under desired level, input parameters (HRT, pH, etc.) can be controlled by this controller, and relevant actions concerning CSTR can be taken.

In this study, a two-layer neural network was used, with a tan-sigmoid (tansig) transfer function at the hidden layer and a linear transfer function (purelin) at the output layer within NN predictive controller (Fig. 3). Tansig is a hyperbolic tangent sigmoid transfer function calculating a layer's output from its net input. $\text{tansig}(N)$ calculates its output according to: $n = 2/(1+\exp(-2*n))-1$. Purelin transfer functions calculate a layer's output from its net input. In the present study, NN has 12 input and one output parameters (Fig. 3).

A. Selection of BP Algorithm

Thirteen BP algorithms were compared to select the best fitting BP algorithm of the data. For all algorithms, a two-layer network, with a tansig transfer function at the hidden layer and a linear transfer function at the output layer, were used. Fifteen neurons were used at the hidden layer. The learning rate parameter may also play an important role in the convergence of the network, depending on application and network architecture. In addition, the momentum allows a network to respond, not only to the local gradient, but also to recent trends in the error surface. Without momentum, a network may get stuck in a shallow local minimum (for details see [12], [26]). In this study, the learning rate and the momentum constant were 0.1 and 0.9, respectively. The training results were as given in Table 2, which are the best performance values (minimum MSE and maximum R) among the ten runs for each thirteen BP algorithms. The best BP

Fig. 3 Optimal neural network structure for prediction of H_2 production ratesTABLE II
PERFORMANCE OF BACK-PROPAGATION ALGORITHMS

Backpropagation Training algorithms (MATLAB function in quotes)	MSE	R	Iteration
Resilient (Rprop) 'trainrp'	0.044	0.915	54
One step secant 'trainoss'	0.081	0.906	40
Powell–Beale conjugate gradient 'traincgb'	0.089	0.904	28
BFGS quasi-Newton 'trainbfg'	0.101	0.903	22
Fletcher–Powell conjugate gradient 'traincgf'	0.096	0.901	27
Gradient descent with momentum and adaptive learning rate 'traingdx'	0.125	0.899	100
Levenberg–Marquardt 'trainlm'	0.014	0.897	10
Scaled conjugate gradient 'traincsg'	0.097	0.896	34
Polak–Ribiere conjugate gradient 'traincgp'	0.067	0.893	41
Gradient descent with adaptive learning rate 'traingda'	0.132	0.889	96
Gradient descent 'traingd'	0.324	0.777	100
Gradient descent with momentum 'traingdm'	0.328	0.757	100
Batch training with weight and bias learning rules 'trainb'	0.424	0.722	100

algorithm, was with a minimum training error (0.0438) and a maximum R (0.92), the Resilient back-propagation (trainrp) algorithm.

B. Optimization of Neural Network Structure

Neuron numbers and relevant performance of the Resilient BP algorithm are shown in Fig. 4. To optimize the neuron number, between 5 to 30 neuron numbers were run ten times with the increments of five. Thereafter, the mean-squared error (MSE), R and iteration numbers were separately evaluated for the neuron numbers. With increasing neuron numbers, MSE decreased for the training set. However, increasing neuron numbers to more than 20 caused an unrealistic result, and a significant change over fitting

occurred. With more than 20 neurons, the mean squared error begins to increase (Fig. 4); therefore, the optimal neuron number for Resilient Back-propagation algorithm is 20.

The optimal neural network structure for the H_2 prediction is given in Fig. 3: a two-layer network, with a tan-sigmoid transfer function at the hidden layer with 20 neurons and a linear transfer function at the output layer. A regression analysis of the network response between the output and the corresponding target was performed. For the output, one regression was determined (Fig. 5).

Taking into account the nonlinear dependency of the data, the output seems to track the targets reasonably well. The R value is 0.91 and the obtained MSE value is 0.085 ± 0.01 . The

training stopped after 30 iterations because the validation error started to increase (Fig. 6).

The performance of the H₂ production rates in CSTR predicted by the model is visualized for experimental data in Fig. 7. There is very good agreement in the trends between forecasted and measured data. This result is reasonable, since the test set error and the validation set error have similar characteristics, and it does not appear that any significant change over fitting has occurred.

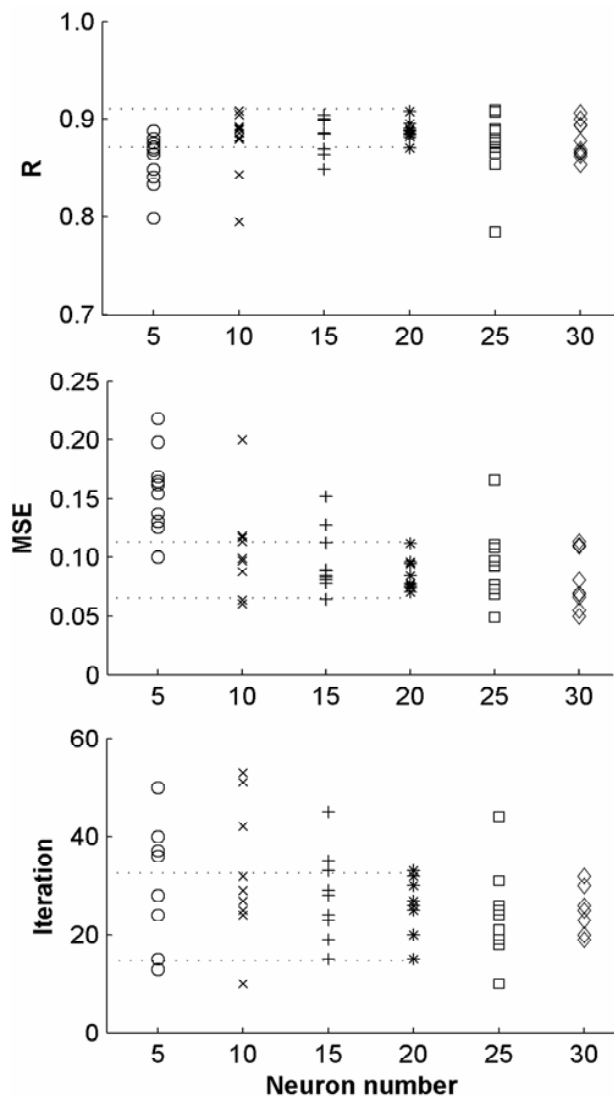


Fig. 4 Neuron number optimization with Resilient (Rprop) ‘trainrp’ BP algorithm. Selected neuron number ‘20’ gives best performance with MSE and R. Trainrp can train any network as long as its weight, net input, and transfer functions have derivative functions. Training stops when any of these conditions occur: The maximum number of epochs (repetitions) is reached; the maximum amount of time has been exceeded; performance has been minimized to the goal; the performance gradient falls below min_grad; and validation performance has increased more than max_fail times since the last time it decreased.

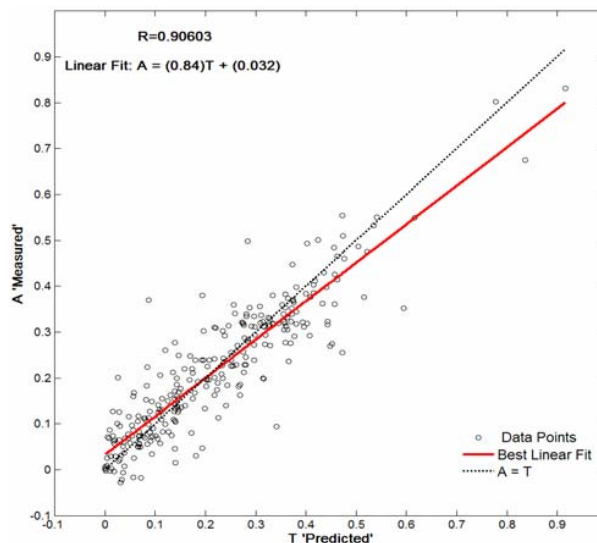


Fig. 5 Linear regression between the network outputs and the corresponding targets for output of Resilient (Rprop) back-propagation algorithm. (A: Measured, T: Predicted)

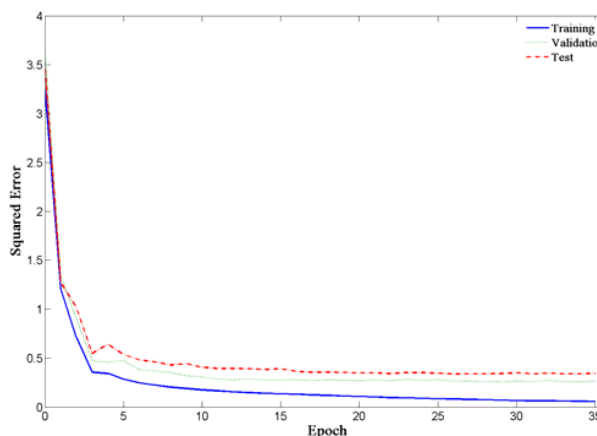


Fig. 6 Training, validation and test mean square errors for Resilient (Rprop) back-propagation algorithm

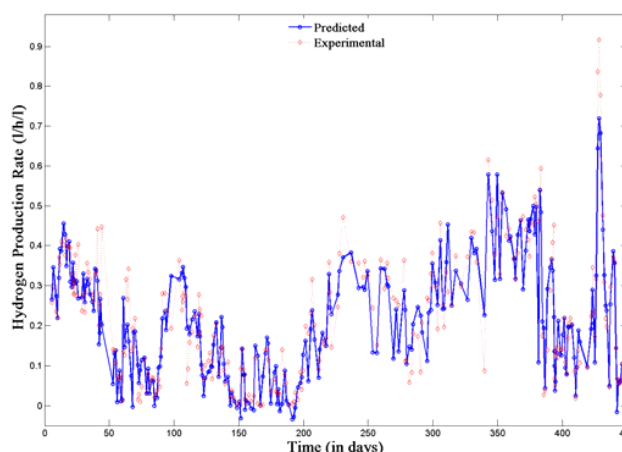


Fig. 7 Predicted and experimentally measured H₂ production rates (solid line: predicted, dashed line: experimental measurements)

IV. CONCLUSION

ANNs can be successfully used to predict H₂ production rate from HRT, recycle ratio, sucrose concentration, sucrose degradation, biomass concentrations, pH, alkalinity, oxidation-reduction potential (ORP), ethanol, acetate, propionate and butyrate concentrations. The ANNs effectively captured nonlinear relationships existing between operational and monitoring variables in a complex multi-input/output system. The proposed ANN based model reliably predicts H₂ production rates and can be used as a predictive controller for management and operation of large scale H₂-fermenting systems.

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REFERENCES

- [1] J'O. M. Bockris, "The origin of ideas on a hydrogen economy and its solution to the decay of the environment," *Int J Hydrogen Energy*, vol. 27, pp. 731-740, 2002.
- [2] D. Das, and T. N. Veziroğlu, "Hydrogen production by biological processes: a survey of literature," *Int J Hydrogen Energy*, vol. 26, pp. 13-28, 2001.
- [3] J. Benemann, "Hydrogen biotechnology: progress and prospects," *Nat Biotechnol*, vol. 14, pp. 1101-1103, 1996.
- [4] I. K. Kapdan, and F. Kargi, "Bio-hydrogen production from waste materials," *Enzyme Microb Tech*, vol. 38, pp. 569-582, 2006.
- [5] C. Li, and H. H. P. Fang, "Fermentative hydrogen production and wastewater and solid wastes by mixed cultures," *Crit Rev Env Sci Technol*, vol. 37, pp. 1-39, 2007.
- [6] R. Nandi, and S. Sengupta, "Microbial production of hydrogen: An overview," *Crit Rev Microbiol*, vol. 24, pp. 61-84, 1998.
- [7] D. B. Levin, L. Pitt, and M. Love, "Biohydrogen production: prospects and limitations to practical application," *Int J Hydrogen Energy*, vol. 29, pp. 173-185, 2004.
- [8] J. Rodriguez, R. Kleerebezem, J. M. Lema, and M. C. van Loosdrecht, "Modeling product formation in anaerobic mixed culture fermentations," *Biotechnol Bioeng*, vol. 93, pp. 592-606, 2006.
- [9] P. Holubar, L. Zani, M. Hager, W. Fröschl, Z. Radak, and R. Braun, "Advanced controlling of anaerobic digestion by means of hierarchical neural networks," *Water Res*, vol. 36, pp. 2582-2588, 2002.
- [10] D. P. B. T. B. Strike, A. M. Domnanovich, L. Zani, R. Braun, and P. Holubar, "Prediction of trace compounds in biogas from anaerobic digestion using the MATLAB neural network toolbox," *Environ Modell Softw*, vol. 20, pp. 803-810, 2005.
- [11] Ö. Cinar, H. Hasar, and C. Kinaci, "Modeling of submerged membrane bioreactor treating cheese whey wastewater by artificial neural network," *J Biotechnol*, vol. 123, pp. 204-209, 2006.
- [12] B. Özkaya, A. Demir, and M. S. Bilgili, "Neural network prediction model for the methane fraction in biogas from field-scale landfill bioreactors," *Environ Modell Softw*, vol. 22(6), pp. 815-822, 2007.
- [13] M. Cakmakci, "Adaptive neuron-fuzzy modeling of anaerobic digestion of primary sedimentation sludge," *Bioprocess Biosyst Eng*, vol. 30, pp. 349-357, 2007.
- [14] E. Sahinkaya, B. Özkaya, A. H. Kaksonen, and J. A. Puhakka, "Neural network prediction of thermophilic (65°C) sulfidogenic fluidized-bed reactor performance for the treatment of metal-containing wastewater," *Biotechnol Bioeng*, vol. 97(4), pp. 780-787, 2007.
- [15] M. Azwar, M. A. Hussain, and K. B. Ramachandran, "The study of neural network-based controller for controlling dissolved oxygen concentration in a sequencing batch reactor," *Bioprocess Biosyst Eng*, vol. 28, pp. 251-265, 2006.
- [16] F. Karaca, and B. Özkaya, "NN-LEAP: A neural network-based model for controlling leachate flow-rate in a municipal solid waste landfill site," *Environ Modell Softw*, vol. 21(8), pp. 1190-1197, 2006.
- [17] B. Özkaya, E. Sahinkaya, P. Nurmi, A. H. Kaksonen, and J. A. Puhakka, "Biologically Fe²⁺ oxidizing fluidized bed reactor performance and controlling of Fe³⁺ recycle during heap bioleaching: an artificial neural network-based model," *Bioprocess Biosyst Eng*, vol. 31(2), pp. 111-117, 2008.
- [18] G. Endo, T. Noike, and J. Matsumoto, "Characteristics of cellulose and glucose decomposition in acidogenic phase of anaerobic digestion," *Proc. Soc. Civ. Engrs*, vol. 325, pp. 61-68, 1982.
- [19] N. Kataoka, A. Miya, and K. Kiriya, "Studies on hydrogen production by continuous culture system of hydrogen-producing anaerobic bacteria," *Water Sci Technol*, vol. 36(6-7), pp. 41-47, 1997.
- [20] C.-C. Chen, and C.-Y. Lin, "Using sucrose as a substrate in an anaerobic hydrogen producing reactor," *Adv Environ Res*, vol. 7, pp. 695-699, 2003.
- [21] C.-Y. Lin, and C.-H. Lay, "Carbon/nitrogen-ratio effect on fermentative hydrogen production by mixed microflora," *Int J Hydrogen Energy*, vol. 29(1), pp. 41-45, 2004.
- [22] C.-Y. Lin, and C.-H. Lay, "Effects of carbonate and phosphate concentrations on hydrogen production using anaerobic sewage microflora," *Int J Hydrogen Energy*, vol. 29(3), pp. 275-81, 2004.
- [23] M. Dubois, K. A. Giles, J. K. Hamilton, P. A. Rebers, and F. Smith, "Colorimetric method for determination of sugars and related substances," *Anal Chem*, vol. 28, pp. 350-356, 1956.
- [24] APHA, *Standard methods*. 19th edn. American Public Health Association, Washington, DC, 1995.
- [25] S. Haykin, *Neural Networks - A Comprehensive Foundation*. New York: Macmillan, 1994.
- [26] M. T. Hagan, H. B. Demuth, and M. H. Beale, *Neural network design*. Boston, MA: PWS Publishing, 1996.
- [27] D. Nguyen, and B. Widrow, "Improving the learning speed of 2-layer neural networks by choosing initial values of the adaptive weights," in *Proc Int Joint Conf Neural Networks*, San Diego, CA, USA, 1990, pp. 321-326.
- [28] H. Abdi, D. Valentin, B. Edelman, and A. J. O'Toole, "A Widrow-Hoff learning rule for generalization of the linear auto-associator," *J Math Psychol*, vol. 40(2), pp. 175-182, 1996.