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NARX Neural Network Modelling of Mushroom Dynamic Vapour Sorption Kinetics

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Abstract: This paper is concerned with the study, optimization and control of the moisture sorption kinetics of agricultural products at temperatures typically found in processing and storage. A nonlinear autoregressive with exogenous inputs (NARX) neural network was developed to predict moisture sorption kinetics and consequently equilibrium moisture contents of shiitake mushrooms (*Lentinula edodes* (Berk.) Pegler) over a wide range of relative humidity and different temperatures. Sorption kinetic data of mushroom caps was separately generated using a continuous, gravimetric dynamic vapour sorption analyser at temperatures of 25-40 °C over a stepwise variation of relative humidity ranging from 0 to 85%. The predictive power of the neural network was based on physical data, namely relative humidity and temperature. The model was fed with a total of 4500 data points by dividing them into three subsets, namely, 70% of the data was used for training, 15% of the data for testing and 15% of the data for validation, randomly selected from the whole dataset. The NARX neural network was capable of precisely simulating equilibrium moisture contents of mushrooms derived from the dynamic vapour sorption kinetic data throughout the entire range of relative humidity.

Keywords: control, optimization, gravimetric, equilibrium moisture content, sorption isotherms.

1. INTRODUCTION

Hygroscopic substances such as agricultural materials and foodstuff are continuously exchanging water vapour with their surrounding environment. Storage stability, physical and mechanical properties are highly affected by moisture content. Growth of microorganisms or quality deterioration may occur if the product's moisture content is higher than a certain value. In general, micro-organisms, such as moulds, yeasts and bacteria increasingly grow at $a_w > 0.7$, while enzymatic activity is also promoted by high values of a_w. Consequently, a threshold a_w much below 0.6 is recommended to prevent quality degradation by microbial or enzymatic activity of dried products during storage. For guaranteeing product safety and quality, the interaction between water molecules and the solid matter of a substance should be controlled. Consequently, the corresponding target moisture content to which the material should be stored or dried can be obtained by the moisture sorption isotherm, which graphically describes the equilibrium relationship between the moisture content (MC) in the sample and the relative humidity (RH) of the atmospheric surroundings at constant temperature and pressure.

In practice, capacitance-type hygrometer sensors or chilled mirror dew point hygrometers represent a fast and robust technique for the indirect measurement of water activity at different temperatures from a common set of partially dried samples (Argyropoulos and Müller, 2014). In addition, vapour pressure manometers are used to measure the actual water vapour pressure exerted by the sample with high accuracy. However, those sorption techniques can not generate kinetic data. For the gravimetric measurement of moisture sorption, the static gravimetric method using thermally stabilized desiccators filled with saturated salt solutions is adopted as a standard (Spiess and Wolf, 1987) while attempts to improve the current experimental procedure have been reported in the literature (Gibert et al., 2006; Wadsö et al., 2004). A climatic test chamber has also been used for the equilibration of samples over specific values of relative humidity and the discontinuous gravimetric measurement on an external balance (Arslan and Toğrul, 2005). In another study, the mass gain due to water vapour adsorption was recorded by placing a balance in the climatic chamber (Stubberud et al., 1995). Instruments using controlled atmosphere microbalances, such as a Dynamic Vapour Sorption apparatus (DVS) have been tested for the automated moisture sorption analysis of pre-dried samples at any desired relative humidity and selected temperature in a comparably short period of time (Argyropoulos et al., 2012; Hill et al., 2010; Kachrimanis et al., 2006; Rocha et al., 2007; Ziegleder et al., 2004).

Arlabosse et al. (2003) published a comparative study between static and dynamic sorption methods and found that thermodynamic equilibrium was dependent on internal diffusion. Modelling of the kinetics of water vapour during adsorption has been mainly performed by the Fick's equation

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(Crank, 1975) based on the diffusion control theory taking into account several assumptions of moisture transfer. However, moisture adsorption is a result of diverse mechanisms besides diffusion such as permeation, bonding at different sites, monolayer and multilayer formation, capillary condensation and evaporation (Kohler et al., 2003). Although several equations have been proposed to fit experimental moisture sorption kinetic data by non-linear regression, neural networks may provide a better approach for optimizing sorption kinetics. An artificial neural network (ANN) can incorporate both linear and nonlinear relationships between input parameters of measured data and network outputs. As a consequence, the application of ANNs has been considerably increased for optimization and control of various engineering and postharvest processes (Boonmung et al., 2006; Brosnan and Sun, 2002; Schulze et al., 2015). In the case of sorption isotherm modelling, ANNs have been used to predict equilibrium moisture contents, obtained by discontinuous gravimetric (Avramidis and Iliadis, 2005; Janjai et al., 2009) and hygrometric (Kingsly and Ileleji, 2009) methods.

On the contrary to those studies that focused on equilibrium, the present work aimed at the sorption kinetics continuously measured by dynamic vapour sorption. Therefore, this paper is concerned with the study, optimization and control of water vapour adsorption kinetics of porous agricultural materials such as mushrooms at temperatures commonly found in processing and storage. It was intended to develop a nonlinear autoregressive neural network capable of precisely simulating equilibrium moisture contents of mushrooms obtained from the dynamic vapour sorption kinetic data throughout the entire range of relative humidity.

2. MATERIALS AND METHODS

2.1 Dynamic vapour sorption

The dynamic, gravimetric vapour sorption experiments were carried out using a DVS-1000 gravimetric moisture sorption analyser (Surface Measurement Systems Ltd., London, U.K.) at the Reutlingen Research Institute, Reutlingen University, Germany. The system consists of a Cahn ultra microbalance with a mass resolution of ± 0.1 µg, two sample crucibles made of quartz and a humidification system in a temperature-controlled incubator. One of the crucibles is used as a reference, whereby the other contains the sample to be measured. A stream of dry and wet nitrogen gas flows along the crucibles. The relative humidity of the mixture is regulated by two electronic mass flow controllers. The apparatus is computer controlled ensuring pre-programming of stepwise variation of relative humidity at set temperature and continuous measurement of temperature, humidity and mass changes during the sorption process.

2.2 Mushroom sample

As porous medium, shiitake mushroom samples (*Lentinula edodes* (Berk.) Pegler) were supplied from a local market in Stuttgart, Germany. Mushrooms were thoroughly cleaned to remove adhering matter, the caps were separated from the stipes and subjected to air drying in a high precision hot-air laboratory dryer (Argyropoulos et al., 2011) until constant mass was achieved. The dried mushroom caps were kept packed in aluminium coated polyethylene bags and stored at ambient conditions in the laboratory before the dynamic vapour sorption trials.

2.3 Experimental procedure

For the DVS experiments, pre-dried samples of shiitake mushroom caps (ca. 10-15 mg) were placed in the quartz sample pan. In order to establish the dry reference mass, each sample was first dried by exposure to dry nitrogen and almost 0% relative humidity until a constant weight of the sample was achieved. The dry reference mass was established in about 960 min. After reaching dry mass, the sample was exposed to stepwise increase of relative humidity, whereby the sample weight was equilibrated at each step. The moisture sorption kinetics was separately determined at 25 and 40 °C by exposing the material to different values of relative humidity ranging from 0 to 85%. Mass gain due to adsorption of water vapour was recorded in two minute time intervals. Temperature and relative humidity data was also measured in close vicinity of the sample. Equilibrium was reached at each relative humidity level by measuring the percent mass change with respect to time (d*m*/d*t*). In particular, equilibrium was considered to have been reached when the change in mass was less than 0.001 mg/min. Once the mass of the sample was below the pre-programmed threshold value, the DVS proceeded to the next target relative humidity step. The values of moisture content at equilibrium, expressed in kg water per kg dry solids (kg/kg d.b.), were used to construct the adsorption isotherms at the examined temperatures. Water activity (a_w) was expressed as equilibrium RH/100.

2.4 Development of the NARX neural network

The nonlinear autoregressive network with exogenous inputs (NARX) is a recurrent dynamic network, with output feedback connections, which enclose several layers of the network. The NARX model is based on the linear ARX model, which is commonly used in time-series modelling. A NARX model can be defined by the following equation (Chen et al., 1990):

$$\begin{aligned} y(t) = f[\,&y(t-1), y(t-2), \ldots, y(t-n_y), \\ &u(t-1), u(t-2), \ldots, u(t-n_u)] \end{aligned} \tag{1}$$

where

$$u = [u_1(t) \quad \cdots \quad u_r(t)]^T, \; y = [y_1(t) \quad \cdots \quad y_m(t)]^T$$

u represent input and y output of the network at time t; n_u and n_y are input and output memory order; and r and m is the number of inputs and outputs, respectively. The function f is a complex function and in this case is approximated by a feed forward neural network. In order to estimate the dynamic

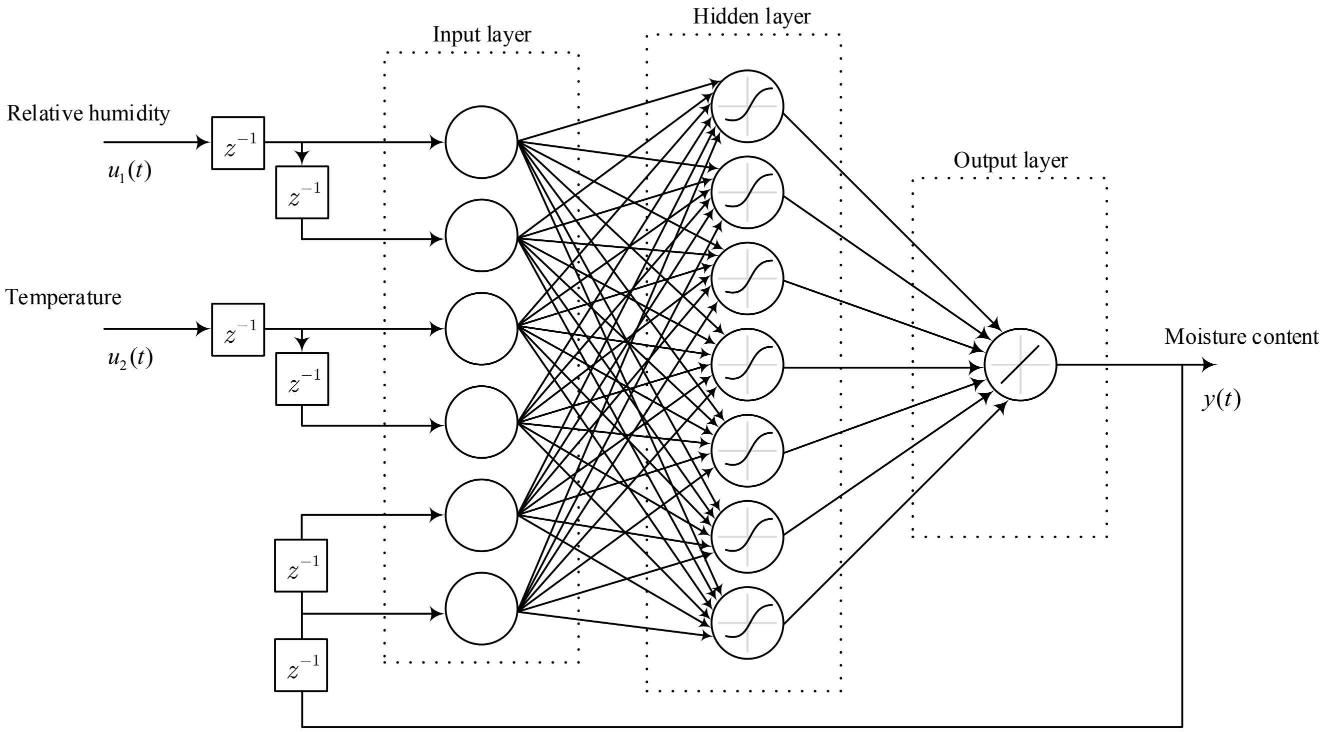

Fig. 1. Architecture of the nonlinear autoregressive network for modelling the dynamic vapour sorption kinetics of mushrooms with two input elements (relative humidity and temperature), one hidden layer and one output layer (moisture content).

vapour sorption kinetics a NARX model was developed using a neural network. The structure of the NARX neural network is schematically shown in Fig. 1. In this study there was only one output (m = 1), namely moisture content and it was modelled using information from relative humidity and temperature (r = 2). The memory order for both n_u and n_y was chosen equal to 2. The input, hidden, and output layer had six, seven, and one perceptrons, respectively. A tangent sigmoid transfer function (tansig) was used for the perceptrons at the hidden layer and a linear transfer function (purelin) for the perceptrons at the output layer.

The neural network was fed with a total sample size of 4500 data points. The development of the NARX neural network involves a series of steps such as the generation of the experimental data required for training, the testing of the network, the evaluation of its configuration resulting in the selection of the optimum and its validation with a data set that was not previously used in training. Hence, the experimental sorption kinetic data was divided into three subsets, 70% of the data were used for training, 15% of the data for testing and 15% of the data for validation. The Levenberg-Marquardt back-propagation method (trainlm) with 500 iterations was selected for training the developed neural network using a training algorithm programmed in MATLAB R2015b (MathWorks Inc., Natick, MA, Neural Network Toolbox™) where several parameters including the learning rule, the transfer function, the learning coefficient ratio, the error minimization algorithm and the number of learning cycles, had to be specified. In the present study various configurations were tested and the network with a seven-node hidden layer was found to have the best performance.

3. RESULTS AND DISCUSSION

3.1 NARX neural network performance

In the current work a NARX neural network was developed to describe the dynamic water vapour sorption kinetics of mushrooms at different temperatures over a wide range of relative humidity as a function of time. A three layer artificial neural network was used for this purpose; the input layer consisted of six neurons represented relative humidity, temperature, time delays and output feedback, while the output layer had one neuron corresponding to moisture content changes during the sorption process. Fig. 2 shows the experimental setup carried out to establish the sorption kinetics of shiitake mushroom caps at 25 °C by dynamic vapour sorption along with the training, validation and testing procedures of the randomly selected data sets. The effect of the increased humidity of air can be observed as an increase in mass of the sample due to water vapour adsorption. The time required for all samples to reach equilibrium was up to 360 min independently of relative humidity level and temperature. Based on the results, the NARX neural network fitted the experimental sorption kinetic data in the entire range of relative humidity sufficiently. For illustration, examples for one of the low (bottom-left) and high (bottom-right) relative humidity steps are also shown in Fig. 2. Fig. 3 shows a comparison between experimental and predicted values of moisture content (kg/kg d.b.) for all test data sets using the NARX neural network. The high correlation coefficient (R^2 = 0.994) indicates that the developed artificial neural network was capable of describing the behaviour of the entire data set with remarkable accuracy.

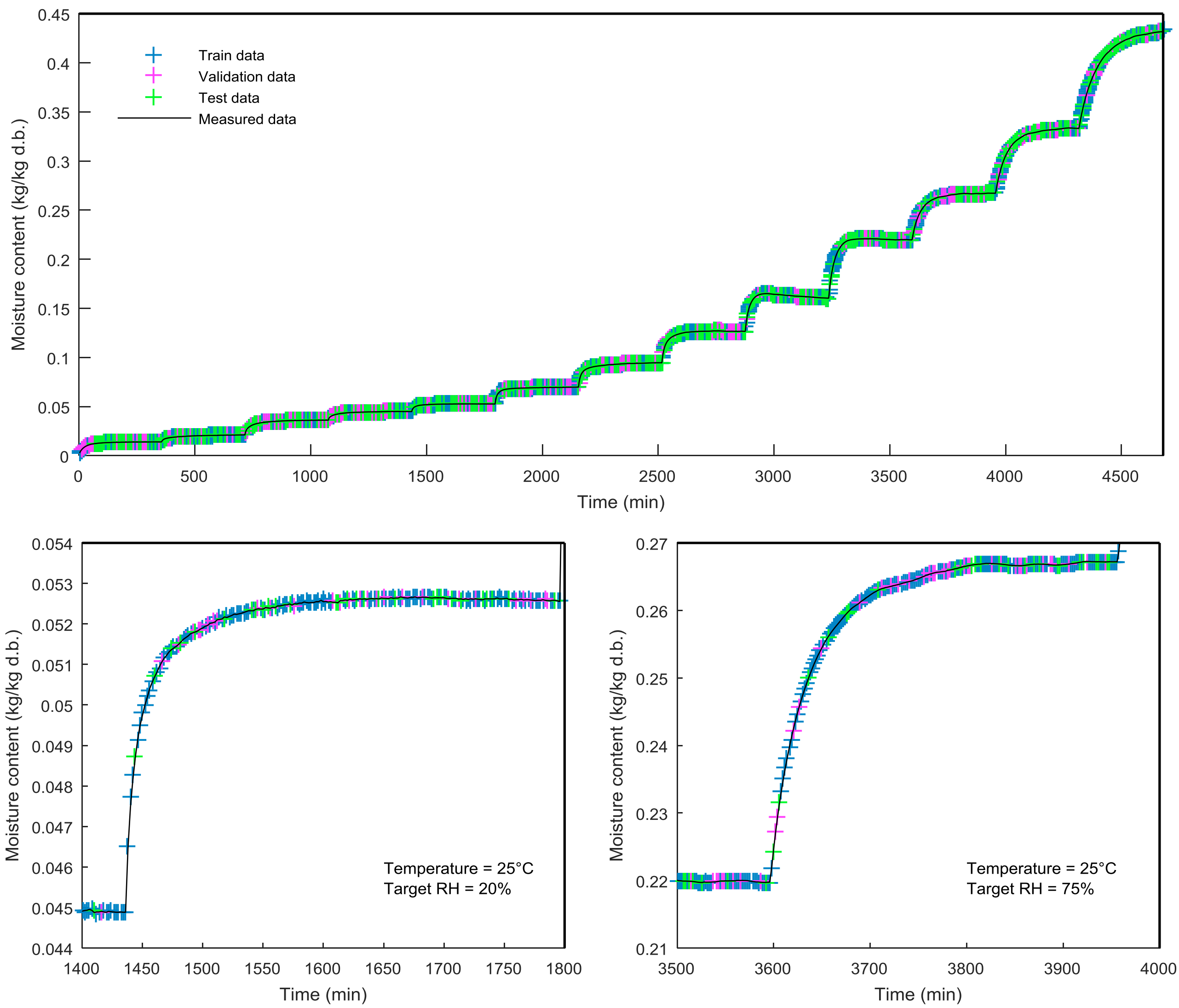

Fig. 2. Training, validation and testing procedures of the NARX neural netowrk in the entire relative humdity range. Low relative humidity level (bottom-left), high relative humidity level (bottom-right).

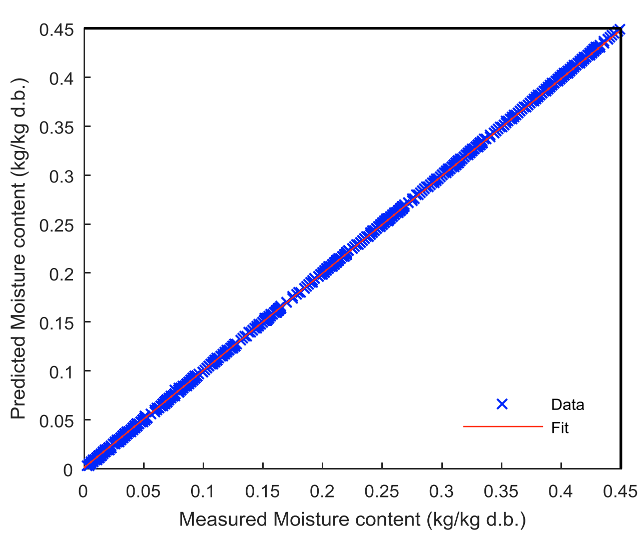

Fig. 3. Comparison of moisture content (kg/kg d.b.) predicted by the NARX neural network with measured moisture content (kg/kg d.b.) established by dynamic vapour sorption.

3.2 Prediction of sorption kinetics by the NARX network

Fig. 4 shows the characteristic changes in moisture content (kg/kg d.b.) of mushrooms as a result of stepwise increase of relative humidity in the DVS versus the process time at constant temperature of 25 °C. The NARX neural network was capable of predicting water vapour sorption kinetics of mushrooms with high accuracy in the entire range of relative humidity tested. Specifically, the mushroom samples were exposed to the following RH profile: 5-20% RH in 5% RH increments, 20-70% RH in 10% RH increments, and 70-85% RH in 5% increments. Nevertheless, the NARX neural network was able to simulate the water vapour sorption kinetics of mushrooms in 1% increments accurately. Particularly, the simulated sorption kinetics of shiitake mushrooms at two different relative humidity levels, low (bottom-left) and high (bottom-right) are also exemplarily shown in Fig. 4. In general, extraordinary good predictions were obtained by the NARX neural network. Less precise simulations were observed at higher relative humidity levels

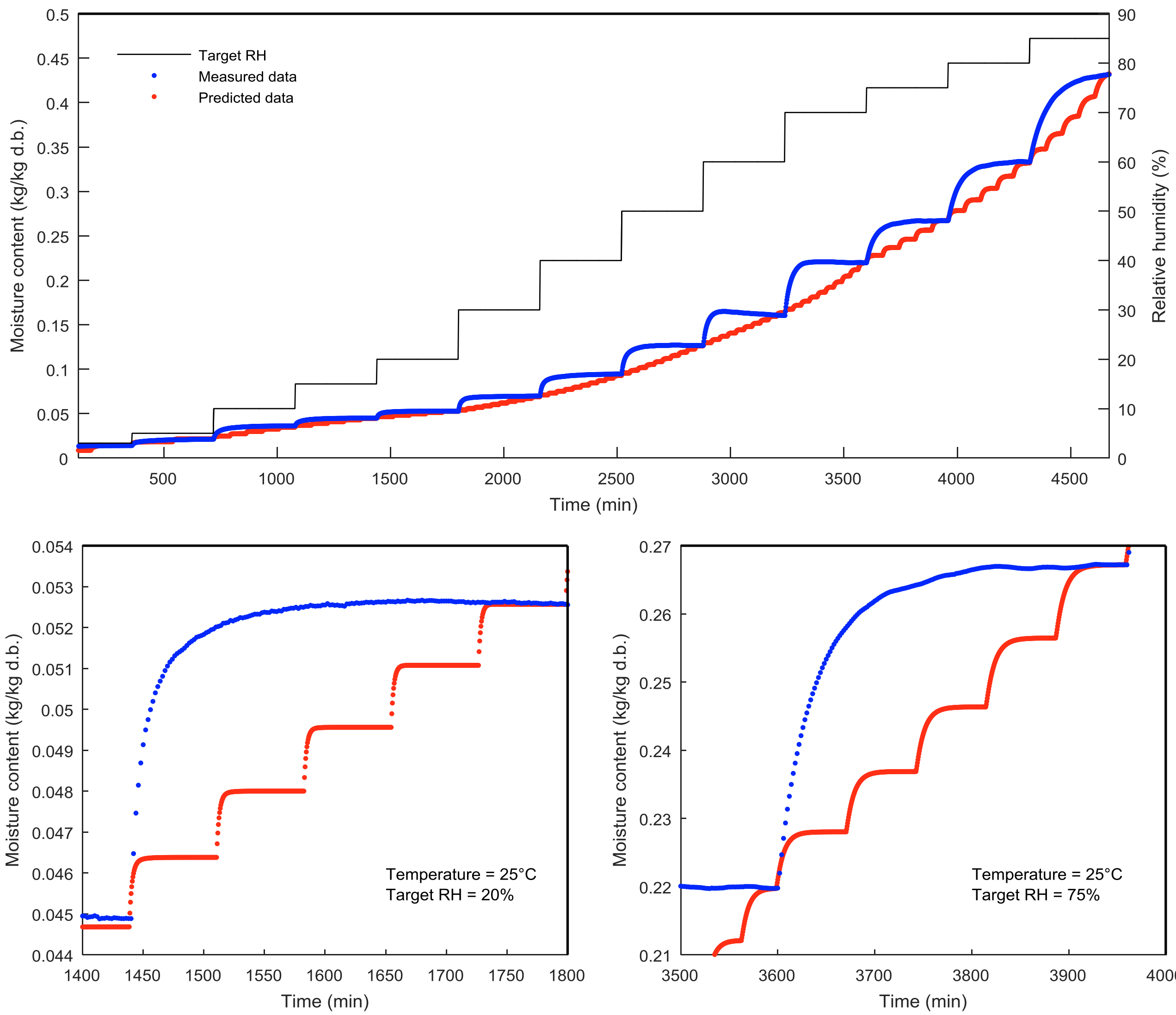

Fig. 4. Predicted water vapour sorption kinetics of mushrooms by the NARX neural network in the entire relative humidity range at 25 °C. Low relative humidity level (bottom-left), high relative humidity level (bottom-right).

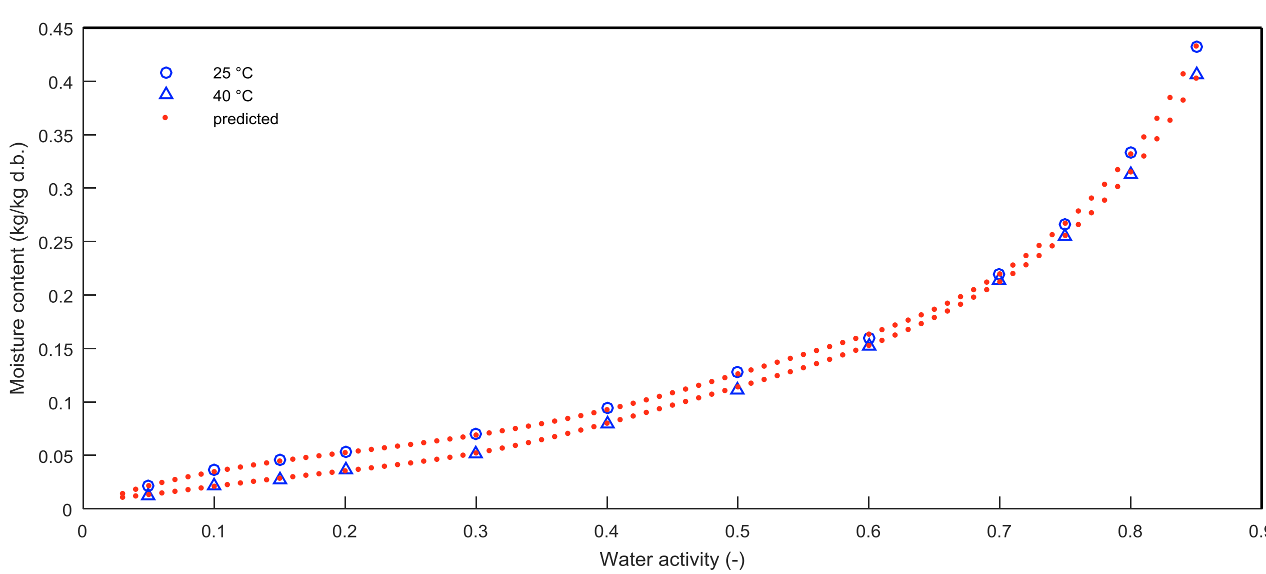

Fig. 5. Equilibrium moisture contents of mushrooms as predicted through the water vapour sorption kinetics by the NARX neural network at different temperatures.

and a temperature of 40 °C presumably due to the larger number of data points measured at equilibrium which increased the experimental noise (results are not shown). The data obtained from the experimental kinetic adsorption profile was further used to calculate the equilibrium moisture contents at the different target values of relative humidity. Moreover, the equilibrium points of the isotherms were also computed from the simulated values of each relative humidity. As shown in Fig. 5, the adsorption isotherms of shiitake mushroom caps were effectively modelled by means of neural networks as a function of water activity and temperature. The obtained isotherms can be used for the optimization of storage conditions in the mushroom industry.

4. CONCLUSIONS

The NARX neural network presented in this study can be applied with high accuracy for estimating equilibrium moisture contents of mushrooms derived from dynamic vapour sorption kinetic data at different temperatures. Modelling of sorption kinetics by neural networks may lead to optimization of equilibrium time for each relative humidity step at specific temperature reducing the time required for a moisture sorption experiment. In contrast to the most of the sorption kinetic models which use physical data for modelling, the developed neural network with the appropriate parameterization would estimate water vapour sorption kinetics taking into account the chemical composition data of the material.

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