NARX Neural Network Modelling of Mushroom Dynamic Vapour Sorption Kinetics

Dimitrios Argyropoulos*, Dimitris S. Paraforos**, Rainer Alex***,
Hans W. Griepentrog**, Joachim Müller*

* University of Hohenheim, Institute of Agricultural Engineering 440e, Garbenstr. 9, D-70599 Stuttgart, Germany
(Tel: +49 711459-23112; e-mail: dimitrios.argyropoulos@uni-hohenheim.de).
** University of Hohenheim, Institute of Agricultural Engineering 440c, Garbenstr. 9, D-70599 Stuttgart, Germany
*** Reutlingen University, Process Analysis and Technology, Alteburgstr. 150, D-72762 Reutlingen, Germany

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

(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 (dm/dt). 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):

$$y(t) = f[\,y(t-1), y(t-2), ..., y(t-n_y), \\ u(t-1), u(t-2), ..., u(t-n_u)] \quad (1)$$

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