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Neural modelling in adsorption column of cholesterol-removal efficiency from milk

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ABSTRACT

In this study, the process of cholesterol removal from milk in an adsorption column with a continuous flow was modelled with artificial neural networks (ANN) models. The input operational parameters used for training the neural network include the bed height (1–3 cm), contact time (0–6 h) and flow-rate (3–9 mL/min). The cholesterol-removal efficiency (%) was defined as the output of the neural network. The neural network structure has been optimised by testing various training algorithms, different number of neurons and activation functions in a hidden layer. A high correlation coefficient (R^2 average ANN = 0.98), a minimum mean-squared error (MSE) and the minimum root mean squared error (RMSE) showed that the neural model obtained was able to predict the cholesterol-removal efficiency in milk. Comparison of the model results and experimental data showed that the ANN model can estimate the behaviour of the cholesterol-removal process through adsorption under different conditions.

1. Introduction

In recent years, the demand for healthy food that is low in fat and cholesterol has increased. In a world of good and bad cholesterol, LDL is the bad one. LDL is called low-density lipoprotein because LDL particles tend to be less dense than other kinds of cholesterol particles. Some LDL cholesterol tends to deposit in the walls of arteries and it is considered to be one of the factors that leads to cardiovascular diseases, which justifies the interest in food with low cholesterol contents (Larsen, 2012). To fulfil this demand, industries have been investing various technologies like supercritical extraction, distillation, enzymatic methods and adsorption techniques. Adsorption, when compared with other techniques, has been presented as an alternative solution for the removal of cholesterol because of its efficiency (Zhang, Zhou, & Lailiang, 2012). The adsorption method, when applied to the removal of cholesterol in milk, has been shown to be complex, because of the numerous constituents present in milk and derived dairy products such as proteins (α-, β-, k-casein), lipids (glycerides and steroids) and sugars (lactose and oligosaccharides) (Oftedal, 2013). This process is multicomponent and suffers from interactions between the large number of variables present (Prakash, Manikandan, & Govindarajan, 2008). Some researches in the food industry make use of phenomenological models in an attempt to describe the adsorption mechanism. Danthine and Blecker (2014) used the Langmuir model to describe the interactions of lipases in milk fat. Silva et al. (2013) used a pseudo-second-order model to describe the adsorption kinetics of carotenes on activated acids. The use of these models requires knowledge of the physical parameters of the process, which often complicate modelling. In response, artificial neural network (ANN) (Aghav, Kumar, & Mukherjee, 2011; Dutta, Parsons, Bhattacharjee, Bandhyopadhyay, & Datta, 2010; Prakash, Manikandan, Govindarajan, & Vijayagopal, 2008; Zoubi, Hilal, Darwish, & Mohammad, 2007) is being considered in the modelling of the adsorption process.

The ANN used here is based non-traditional tools for modelling the adsorption process. The success of this ANN modelling technique in adsorption processes has been attributed to the powerful mathematical tools of ANNs, which is capable of modelling complex and non-linear systems. Instead, it uses process data to predict the relationship between input and output parameters (Kashaninejad, Dehghani, & Kashiri, 2009; Kumar & Porkodi, 2009).

The progress of empirical models obtained from numerical estimation techniques based on ANN has made this tool an alternative to predict adsorption systems. According to previously published reports, numerous studies have been conducted to predict performance in the removal process through adsorption when

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using ANN modelling (e.g. Asl, Ahmadi, Ghiasvand, Tardast, & Katal, 2013; Kumar & Porkodi, 2009; Turan, Mesci, & Ozgonenel, 2011). Asl et al. (2013) used ANN to investigate the percentage of chromium Cr (VI) removal during an adsorption process. Using feed-forward architecture and the backpropagation algorithm, the model presented a mean square error (MSE) of 0.0002 and a correlation coefficient of 0.98. Kumar and Porkodi (2009) used an ANN to analyse the second-order kinetics of the adsorption of the auramine dye process. The results showed that the neural model has a correlation coefficient of 0.99 and a low MSE (denormalized data MSE = 7.23) relative to the experimental data. Turan et al. (2011) used ANN in order to optimise the removal efficiency of Zn (II) ions in the adsorption process, using hazelnuts as a biosorbent. The results show that the proposed model was capable of predicting the level of Zn removal with a correlation coefficient of 0.99 for a maximum removal of 87.52%. However, neural modelling of the adsorption of cholesterol in milk could not be found in previously published studies.

In this work, the effect of operating parameters such as flow rate (mL/min), contact time (h) and column height (cm/amount of adsorbent) on the adsorption process was investigated in an adsorption column filled with an adsorbent that was obtained by the molecular imprinting of the biomolecule of interest. The ANN technique was used to process the modelling efficiency of cholesterol removal in milk. The influence of each experimental parameter was investigated using the predictive ability of the ANN model. An explanation of the simulated results from the models and the predicted results from ANN are presented.

2. Materials and methods

2.1. Milk

The samples used in the process were of cow's milk (3–5 percent fat).

2.2. Reagents

The used reagents were tetraethyl orthosilicate (98%), acetonitrile (99.9%), isopropanol (99.7%), hexane (65%), hydrogen peroxide (32%), sodium hydroxide (97%), potassium hydroxide (85%), hydrochloric acid (37%), ammonium hydroxide (30%), chloroform (99.8%) and ethanol (99.8%). All reagents used were of analytical grade.

2.3. Adsorbent preparation

The methodology for the preparation of the molecularly printed adsorbent was based on the procedure described by Soares et al. (2011). An aliquot of tetraethyl orthosilicate (30 mL) was dissolved in absolute ethanol (36 mL) under an inert nitrogen atmosphere. In the next step, diluted hydrochloric acid (0.11 mL) in ultrapure water pre-hydrolysis solution (5 mL) was added and the mixture was stirred (20 rpm) at 35 °C for 90 min. After the repose period was reached, the cholesterol (0.51 g) dissolved in ethanol (10 mL) and dilute ammonium hydroxide (1 mL) in ethanol hydrolysis solution (6 mL) was added to the mixture, and this was then left for an additional 30 min without agitation at a constant temperature until gelification occurred. After gelification occurred, the mixture was cooled and allowed to stand for 18 h at 4 °C for polycondensation to occur. The obtained gel was washed with ethanol and water at 50 °C and dried under vacuum at room temperature. It was then triturated, giving a powder with a 250 μm particle size.

2.4. Experimental study

2.4.1. Adsorption in the column

The experiments were performed in a continuous stream through an adsorption column jacketed with glass with an inner diameter of 1 cm and a height of 10 cm, using a peristaltic pump to control the flow (Soares, Zanin, Moraes, Santos, & Castro, 2007) as shown in the experimental diagram in Fig. 1.

The adsorption column was jacketed at 42 °C and filled manually with silica mass, through which a continuous flow of the milk sample, controlled by the peristaltic pump, was injected. At the base of the column, glass wool was used to prevent small particles of the adsorbent passing to other parts of the experimental unit. To determine the concentration, fractions of the output mobile phase of the layer were collected in a 200 mL amber glass at time 0, over a period of 5 min starting after 15 min, over a period of 15 min starting after 90 min, over a period of 30 min after 4 h, and then every hour until 6 h of removal was reached. The samples were collected and analysed by HPLC. The percentage cholesterol removal was the output parameter of the ANN model, which was considered to be a measure of the adsorption efficiency (R%) of the column. The efficiency of adsorption was calculated as follows (Eq. (1)):

$$R(\%) = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (1)$$

in which R(%) is the cholesterol-removal efficiency, C_0 is the initial concentration and C_e is the equilibrium concentration.

2.4.2. Analytical determinations

Samples were derivatised before chromatographic analysis. For such analysis, the sample (10 g) was weighed and a solution containing KOH and ethanol was added (10 mL in a 1:9 M ratio); this was left for 10 min at 60 °C before distilled water (10 mL) was added. After cooling the sample, the supernatant was extracted with n-hexane (15 mL). After separating the phases, the sample was dried under an inert atmosphere. The sample was reconstituted in a mobile phase constituting acetonitrile and isopropanol in a 9:1 M ratio. Immediately after reconstitution, the Pro Star (Varian) chromatograph, UV/Vis detector (SPD-10 AVvp) and software (Varian Star Integrator Program version 4.5) were use to obtain the data. The analytical column was a C18 column (150 × 4.6 mm × 5 μm; Gemini, Phenomenex), which was maintained at 40 °C in a temperature-controlled oven. The derivatised samples were injected into the chromatograph. The mobile phase was composed of 90% acetonitrile and 10% isopropanol, administered at a flow rate of 1.5 mL/min with an analysis time of 20 min. Chromatograms were obtained at a wavelength of 210 nm.

2.5. Definition of ANN models

In this study, in order to model cholesterol adsorption efficiency with ANN, the NNTOOL (neural network toolbox) of MATLAB

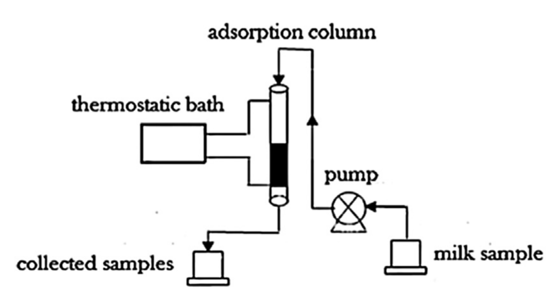

Fig. 1. Experimental unit for the removal of cholesterol in milk.