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Estimation of the storage properties of rapeseeds using an artificial neural network

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ABSTRACT

Rapeseed losses during storage can lead to undesirable difficulties in oil and biodiesel production. In this paper, three artificial neural networks were created to anticipate the main quality parameters of thirteen rapeseed varieties - cultivars and hybrids (*Brassica napus* L.) - during drying and storage. The varieties, drying temperature, air velocity and drying time were used as inputs to the artificial neural network model to predict the changes in seed weight and moisture during the drying process. The moisture diffusivity and activation energy of the investigated rapeseed varieties were determined under convective drying. For the experiment, an on-site drying system was used at 40, 60 and 80 °C drying air temperature. The effective diffusivity ranged from: $7.947.10^{-10}$ to $1.459.10^{-8}$ m^2/s (first drying period) and $4.716.10^{-10}$ to $8.611.10^{-9}$ m^2/s (second drying period). The predicted Arrhenius constant and activation energy ranged from 17.169 to 42.546 kJ/mol (first drying period) and from 31.261 to 50.474 kJ/mol (second drying period). Seed oil content, free fatty acids and thousand seed weight were determined after drying at different temperatures and after 12 months of storage under the three different storage conditions. To predict these parameters after storage time, a multilayer perceptron model with three layers (input, hidden and output) for three artificial neural networks (ANNs) was used for modelling using the implemented drying parameters (such as: variety, drying temperature, air velocity and drying time, along with initial oil and free fatty acid content and storage type) were used. The prediction of the developed model was accurate enough for the prediction of the output parameters. The coefficients of determination ranged from 0.965 to 0.998 when predicting the weight and moisture of the rapeseed during the drying process and the oil and free fatty acid content and thousand grain weights after the 12 months storage period.

1. Introduction

Rapeseed (*Brassica napus* L.) is an important oil crop grown mainly in the European Union, China, Canada, India and Australia. It is the third-largest source of vegetable oil in the world after soybean and palm oil (USDA, 2015). Furthermore, rapeseed provides a significant part of the world's vegetable oil production; about 44% of the seed dry matter is oil (Wittkop et al., 2009). Rapeseed plays an essential role in global agriculture and is important for industrial applications. In the food industry, it is one of the most important raw materials and its advantages derive, among other things, from the fact that it contains a high proportion of oil, which is characterised by a favourable fatty acid composition. The most abundant fatty acids are linolenic acid (C18:3), linoleic acid (C18:2), oleic acid (C18:1) and erucic acid (C22:1) (Gawrysiak-Witulska et al., 2011; Siger et al., 2018; Wawrzyniak et al., 2019). Other classes of compounds include tocopherol (vitamin E), cellulose, phenolic acids and glucosinolates. Rapeseed oil is also a rich source of bioactive components with health-promoting properties, which opens up the possibility of its use in the production of functional foods. Sterols, tocopherols and phenolic compounds in rapeseed are bioactive components that have antioxidant effects (Gawrysiak-Witulska et al., 2012). Rapeseed oil is extracted from the seeds for the production of edible oil, margarine and salad dressings in the food industry, and the protein-rich by-product is used as a high-protein feed meal in livestock farming (Nogala Kalucka and Siger, 2010). Not only is the oil a component of a healthy human diet, but rapeseed meal or cake residues (after oil extraction) are also a protein-rich feed ingredient. Rapeseed is the most important raw material for biodiesel production (Niedbała, 2019).

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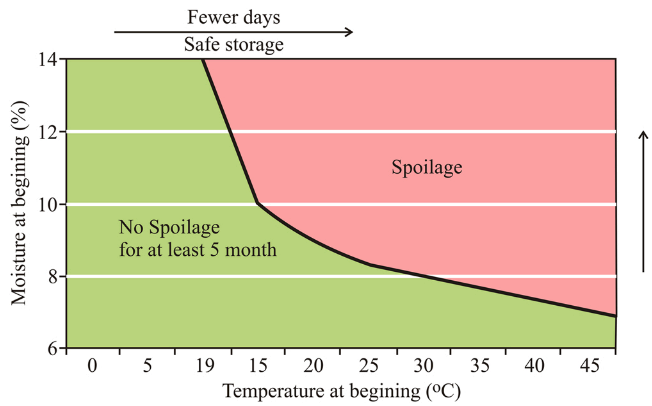

Fig. 1. Conditions in which seed spoilage occurs in the warehouse (www. canola encyclopaedia).

The development of improved rapeseed varieties, i.e. free of harmful erucic acid and with a reduced glucosinolate content (the so-called double zero "00" varieties), has increased interest in this crop in recent decades. As a result, rapeseed cultivation has increased significantly and global production has risen on average from 10.8 million tonnes in 1980–75.0 million tonnes in 2018. In recent years (2009–2018), the largest rapeseed growers were Canada (24.6 %), China (13.7 %), India (7.4%), France (5.1 %), Germany (7.3 %), Australia (4.7 %), Poland (3.5 %) and the UK (3.3 %). In the EU, 25.49 million tonnes of rapeseed were produced in 2018 (FAO, 2021).

Increasing rapeseed production requires appropriate postharvest processes to minimise seed deterioration and ensure food safety. Rapeseed drying is a critical step (Duc and Han, 2009) or one of the most important steps in the postharvest or "post-cropping" process (Nogala Kalucka and Siger, 2010). Factors that influence the quality of rapeseed during drying are: air temperature and initial seed moisture content, drying time and air velocity (Fornal et al., 1994; Tys et al., 2002; Laoretani et al., 2014; Gawrysiak-Witulska et al., 2016).

The drying time depends on the air temperature at which the seed is dried and the air velocity in the dryer. The higher the temperature of the air used to dry the seed, the shorter the drying process (Laoretani et al., 2014) or as the air temperature increases, the water release from the seed increases (Jukić et al., 2009). The choice of air temperature for drying the seeds also depends on the moisture of the seeds before drying. According to previous research, the air temperature used to dry the seed must be lower at the beginning when the seed moisture content is higher. As the drying process progresses, the temperature can then be increased without major losses in quality (Kanai et al., 2010). It is extremely important to ensure that the temperature of the seed does not exceed certain limits, after which irreversible quality changes occur. These facts were confirmed by Laoretani et al. (2014), who dried seeds of an rapeseed variety with air at temperatures of 35, 60, 82 and 100 °C. The moisture content of the seeds before drying was 13.6 % and 22.7 %. Samples whose moisture content before drying was 22.7 % showed a reduction in tocopherol content after drying. The only exception to this rule was when the seeds were dried with air at a temperature of 35 °C. Samples of seeds with a moisture content of 13.6% before drying showed no reduction in tocopherol content, regardless of the air temperature at which the seeds were dried.

Because of its morphology, structure and chemical composition, rapeseed is not easy to store until the moment of processing. Namely, during the storage of rapeseed, factors occur that affect the quality of the seeds and the oil. Rapeseed must be cleaned immediately after harvest and, if necessary, dried below the critical moisture content. The moisture content of the seeds in Croatia, as in all areas in SE Europe, is between 7% and 15 % at harvest. The critical moisture content of rapeseed is between 9 % and 10 % (Jukić et al., 2009). Some authors indicate the moisture content at which rapeseed can be stored. According to Gawrysiak-Witulska et al. (2012), the recommended moisture content for seed storage is between 7 % and 10 %. Duc and Han (2009) state that rapeseed must be dried after harvest to reach a final moisture content of 8 % (d.b.), which is ideal for safe storage. The safe storage period depends on seed temperature, seed moisture content and seed oil content (Banks, 1998). Seed temperature and moisture content at the beginning and duration of storage are the most important factors affecting fungal development in stored rapeseed (Mills and Sinha, 1980). If the seed is to be preserved over a long period of time without major losses, the moisture content of the seed must be low and around 6 %, as shown in the following figure (www.canola encyclopaedia).

Fig. 1.

The quality parameters of rapeseed are the oil and fatty acid content, but also other bioactive components in the seed such as lipids, tocopherols, sterols, carotenoids and phenolic compounds (Nogala Kalucka and Siger, 2010). The same authors found in their research that the most unfavourable influence on native hydrophilic antioxidant content was observed after drying at 60 and 80 °C. Seeds dried at these temperatures are characterised by the greatest decrease in total phenolic compounds and free phenolic acid content. According to Tys et al. (2002), pigment content in both seed and oil is a significant indicator of oil quality, especially for cold-pressed oil.

Increasing the temperature during the drying of rapeseed has a negative effect on the colour of the oil. The most detrimental changes occurred in seeds dried at a temperature above 120 °C, most probably due to the formation of free radicals, Tys et al. (2002).

Drying also affects the occurrence of seed breakage and oil yield during seed processing (Sadowska et al., 1995), the number of broken seeds also has a negative impact on the colour of the oil, Tys et al. (2002). Fornal et al. (1994) state that the poor processing quality of rapeseed results from the changes in mechanical properties resulting from microstructural changes in the seed due to improper postharvest treatment. According to previous authors, the changes in the microstructure of rapeseed are strongly influenced by heat treatment during postharvest drying. Initial seed moisture combined with high temperature has a stronger effect than temperature alone. Greater susceptibility of rapeseed to crushing and lower oil recovery during extraction could be the result of improper postharvest drying. Kanai et al. (2010) find that early harvested rapeseed dried under higher temperature conditions not only exhibited oil quality as measured by peroxide value but also green colour, which is due to chlorophyll. During the drying process, air temperature and relative humidity are two important factors affecting seed germination characteristics (Duc and Han, 2009). Faron and Tańska (2013) indicate that rapeseed with a moisture content of 11% should be dried no later than 4 days after harvest and seeds with a moisture content of 17% no later than 2 days after harvest.

Therefore, modern agricultural postharvest management aims to find an effective control system for maintaining the characteristics of the processed seed. Until recently, the prediction of properties during postharvest processing has mainly focused on mathematical models describing the effects of environmental factors on processed seed quality. There are numerous kinetic models in the literature that represent the changes in seed persistence under certain circumstances (primary models), models that represent the relationships of the parameters of the primary model to ecological variables (secondary models), and models that combine these two approaches (tertiary models) and describe the dynamic conditions of seed changes (Marks, 2008; Natskoulis and Panagou, 2020; Wawrzyniak, 2020; Rajković et al., 2022). Artificial neural networks (ANN) are widely used to represent nonlinear systems and are an interesting alternative to existing kinetic models of quality parameter change during storage. Therefore, ANN represents a promising modelling approach that can also be used to control the process during the storage of rapeseed. Wawrzyniak (2020) investigated the ability of ANN based on multilayer perceptrons and radial basis functions to anticipate the quality parameters of stored rapeseed at different

Table 1
Rapeseed properties during harvesting at 7 % moisture.

Variety	Moisture content during harvesting (%)	Seed yield (kg/ha)	Oil yield (kg/ha)
1	16.9 ± 1.7 ^{cd}	3058 ± 282^{d}	1.312 ± 130^{bc}
2	10.2 ± 0.9^{a}	3525 ± 359de	1.473 ± 162^{bc}
3	17.2 ± 1.6^{d}	3124 ± 332^{d}	1.337 ± 136^{bc}
4	18.6 ± 1.8^{e}	2135 ± 233^{b}	908 ± 83^{b}
5	15.7 ± 1.5^{c}	3605 ± 332de	1.579 ± 148^{c}
6	17.6 ± 1.6^{d}	3523 ± 344de	1.622 ± 150^{c}
7	14.8 ± 1.4^{b}	2530 ± 269^{c}	1.044 ± 95^{b}
8	9.8 ± 1.1^{a}	3430 ± 351de	1.525 ± 166^{c}
9	13.7 ± 1.5^{b}	2667 ± 244^{c}	1.221 ± 117^{bc}
10	16.4 ± 1.6 ^{cd}	1690 ± 165^{a}	692 ± 76^{a}
11	18.2 ± 2.0^{de}	4180 ± 426^{e}	1.719 ± 156^{d}
12	15.4 ± 1.4^{c}	3960 ± 424^{e}	1.715 ± 187^{d}
13	16.4 ± 1.6 ^{cd}	3825 ± 396^{e}	1.585 ± 156^{c}

The means in the same column with different superscript letters are statistically different ($p \leq 0.05$), according to Tukey's HSD test

Table 2
The obtained yields of investigated cultivars of rapeseed.

Cultivar	Oil (%)		FFA (%)		TKW (g)	
	Before drying	After drying	Before drying	After drying	Before drying	After drying
1	43.2 ± 0.8^{d}	40.1 ± 1.4^{ab}	1.7 ± 0.4 ^{g}	2.1 ± 0.5 ^{f}	5.7 ± 0.3 ^{cd}	5.4 ± 0.5 ^{cd}
2	42. 5 ± 0.7^{c}	39.6 ± 1.1^{a}	2.3 ± 0.4 ^{h}	2.6 ± 0.6 ^{g}	5.4 ± 0.4^{c}	5.1 ± 0.1^{c}
3	44.0 ± 0.8^{e}	41.1 ± 1.0^{bcd}	1.3 ± 0.2^{def}	1.5 ± 0.3^{bcde}	5.6 ± 0.8^{c}	5.2 ± 0.4^{c}
4	42.5 ± 0.2^{c}	41.3 ± 0.8 ^{cd}	1.1 ± 0.2^{bcd}	1.3 ± 0.2^{bc}	3.9 ± 0.1^{ab}	3.6 ± 0.2^{ab}
5	42.9 ± 0.6 ^{cd}	42.5 ± 0.4^{ef}	1.4 ± 0.0^{ef}	1.7 ± 0.2^{cde}	3.2 ± 0.4^{ab}	3.0 ± 0.5^{ab}
6	46. 0 ± 0.1 ^{f}	45.2 ± 0.5 ^{g}	1.2 ± 0.0^{cde}	1.4 ± 0.1^{bcd}	3.1 ± 0.3^{a}	2.8 ± 0.4^{a}
7	41.2 ± 0.1^{a}	40.3 ± 0.5^{abc}	1.5 ± 0.0 ^{f}	1.8 ± 0.2^{ef}	3.2 ± 0.6^{ab}	3.0 ± 0.5^{ab}
8	44.5 ± 0.0^{e}	43.6 ± 0.3 ^{f}	1.4 ± 0.0^{ef}	1.8 ± 0.2^{de}	4.2 ± 0.7^{b}	3.9 ± 1.6^{b}
9	45.8 ± 0.0 ^{f}	44.8 ± 0.6 ^{g}	1.4 ± 0.0^{ef}	1.7 ± 0.2^{cde}	2.9 ± 0.3^{a}	2.7 ± 0.2^{a}
10	43.0 ± 0.0 ^{cd}	42.0 ± 0.7^{de}	0.6 ± 0.0^{a}	1.1 ± 0.1^{ab}	3.5 ± 0.7^{ab}	3.2 ± 1.2^{ab}
11	41.2 ± 0.1^{a}	40.3 ± 0.8^{abc}	0.6 ± 0.0^{a}	0.9 ± 0.2^{a}	6.9 ± 1.2^{d}	6.3 ± 1.0^{d}
12	43.3 ± 0.0^{d}	41.1 ± 0.8^{bcd}	1.0 ± 0.0^{b}	1.3 ± 0.1^{ab}	6.0 ± 0.5 ^{cd}	5.8 ± 0.6 ^{cd}
13	41.8 ± 0.3^{b}	40.1 ± 1.2^{ab}	1.0 ± 0.0^{bc}	1.2 ± 0.1^{ab}	6.6 ± 1.2^{c}	6.3 ± 1.0^{c}

The means in the same column with different superscript letters are statistically different ($p \leq 0.05$), according to Tukey's FFA - free fatty acids content (g), TSW – 1000-seed weight

temperatures and water activities in the seeds. During the development of the ANN model, different configurations of ANN were tested, using different numbers of hidden layers, different numbers of neurons and activation functions in hidden and output layers. The chosen activation function in the hidden layer of the optimal ANN network was the hyperbolic tangent function, while the identity function was the activation function in the output layer. The created ANN structure had high prediction accuracy and high generalisation capacity, which showed that the result of the ANN model can be used for prediction as a supporting tool in the research of quantifiable parameters in the context of post-harvest storage control of rapeseed.

However, it is crucial to emphasise that even the best modelling methods are not convincing if they are not based on accurate data. An important prerequisite for the development of predictive tools for postharvest planning is data from experiments conducted in an environment that is as close as possible to real systems. The lack of objective information covering the full range of conditions is one of the critical shortcomings in the further development of such tools (Marks, 2008). Although mathematical models for individual drying and storage analysis of rapeseed are often described in the literature, these are based on data obtained under ideal laboratory conditions. Indeed, maintaining the seed quality of rapeseed is a priority in cultivation. Nagel et al. (2018) have shown that the complexity of seed longevity was revealed by a unique viability data set obtained during long-term storage. It was hypothesised that in addition to environmental factors, seed composition also influences seed viability after long-term storage. Linear regression was found to support this hypothesis, but the coefficients were too low to provide reliable interpretations. In contrast, multivariate approaches based on machine learning and ANN simultaneously analyse the effects of multiple parameters to reveal some key seed components such as fatty acids, oil content and glucosinolates that influence seed viability (Nagel et al., 2018). According to Rajković et al. (2021), it is possible to predict the quality of rapeseed based on oil and protein content, seed yield, oil and protein yield and 1000-kernel weight (TKW), based on year of production and genotype, using ANN and random forest regression (RFR) models.

Accordingly, the main purpose of this investigation was to develop ANN models for predicting changes in some quality parameters of the most important quality properties of the thirteen Croatian rapeseed varieties (cultivars and hybrids) used for biodiesel production during 12 months of storage. Variety as a categorical variable and drying temperatures were chosen as input variables and implemented as variables in all three ANN models. In the first ANN model (ANN1), air velocity and drying time were also used to model seed weight and seed moisture content during the drying treatment. The ANN2 model was used to predict the oil and free fatty acid content (FFA) after the 12 months of storage period as a function of the initial oil and FFA content and the drying method. Finally, the third model ANN (ANN3) predicted the thousand-seed weight after the one-year storage period depending on the drying type.

2. Materials and methods

2.1. Laboratory analysis of drying and storing of rapeseed

Immediately after the harvest, seed samples of 13 rapeseed varieties were delivered to the Faculty of Agriculture at the University of Zagreb. During harvesting, seed properties and seed and oil yields (kg/ha) were determined. Table 1. shows the characteristics of the rape during harvest.

In the laboratory, the samples were cleaned of impurities and then stored in a room near the laboratory at low temperature and relative humidity. The moisture content of the seed was determined before drying and before storing the seed samples according to the standards of ISO (HRN EN ISO 665:2004). After measuring the initial moisture, oil and FFA content, the drying process was continued to a value of 6% of the average moisture.

The determination of water activity of saturated salt solutions was discussed in the investigation of Corey et al. (2011) and Pedrali et al. (2020). The samples were brought to equilibrium, at which their moisture content was determined after drying in a vacuum oven at 70 °C and 50 torr for 18 h. The determined plots of moisture content vs. water activity were fit to the Guggenheim-Anderson-de Boer model (Labuza and Altunakar, 2007). Within this research the final product is not considered to be consumed as a food ingredient, but instead to be used for biodiesel production. Therefore, the oxidation phenomenon was not considered and peroxide number was not examined (it was not contemplated as relevant for this research) and also the water activity was not determined.

The drying process was performed at temperatures 40, 60, and 80 °C, during which oxidation of FFA did not occur, as shown in Table 2. The results shown in Table 2 indicate that the FFA content increases during the drying period, due to moisture loss (hence the relatively low temperatures did not affect FFA mass transfer).

Drying was carried out in a laboratory dryer. The seeds were dried at the following temperatures: 40, 60 and 80 °C. The velocity of the air flowing through the seed layer was between 1.0 and 1.6 m/s. Drying was continued until the moisture content of the seed reached 6 %. Similar drying temperatures were also used by other authors (Duc and Han, 2009).

The drying process began when the target input temperature and air velocity were reached by the seed layer. The seed mass was measured at 5-minute intervals from the beginning to the end of drying. The dryer consists of a power generating part, an operating part and a control part. The power-generating part consists of an electric motor (which supplies air via a ventilator), electric heaters and a copper hull. The active part consists of a borosilicate cylinder and two separating grids that provide optimal air distribution for drying through the layer of dry material. The control part allows the electrical potential to be changed using an autotransformer by regulating the speed of the electric motor. The operating range of the control is between 40 °C and 150 °C. The air temperature was measured with a PT1000 probe with an accuracy of ± 0.35 °C. The air velocity was measured with a digital anemometer with an operating range of 0.3–30 m/s. All weight measurements, as well as the weight of one thousand seeds, were made using a digital balance with two decimal places.

After drying in the seed samples, the grain moisture, oil content and free fatty acid content were determined. The oil content was determined according to HR EN ISO 659: 2009 and the free fatty acid content according to HR EN ISO 729: 1998. Also, after drying, the seed samples were stored under three different temperature-humidity regimes (at room temperature and under controlled conditions at 4 °C and 10 °C). Thousand seed weight, oil content and free fatty acid content were determined at the beginning and end of the twelve (12) months storage. The water content was determined according to the method HR EN ISO 665: 2020, the oil content according to the method HR EN ISO 659: 2009 and the free fatty acid content (FFA) according to the method ISO 729:1988.

2.2. Mathematical model of drying curve

The moisture ratio of the rapeseed (MR) throughout the drying process was modelled using the following equation (Doymaz, 2012; Šobot et al., 2019):

$$MR = \frac{M - M_e}{M_0 - M_e} \tag{1}$$

where: M_0, M, M_e - moisture content at the beginning of the process, at any time, and equilibrium moisture content, respectively.

According to the theory of drying (Doymaz, 2012), the drying curve in the product is defined as a function of time in which there are two periods: an interval of constant rate (linear period of the drying curve) followed by a period of falling rate. The constant rate period is considered much faster, while the falling rate period is slower, takes longer and the residual moisture is then controlled by diffusion. The constant rate period is calculated because it is an almost externally controlled phase, influenced only by the drying conditions (drying time and temperature) but not by the product characteristics.

The experimental results were evaluated according to diffusivity coefficients, upon Fick's second diffusion model (Doymaz, 2012):

$$\frac{\partial M}{\partial t} = D_{eff} \cdot \nabla^2 M \tag{2}$$

The computation of the moisture gradient was considered one-dimensional, and the analytical solution of Eq. (2) was elucidated, assuming that the temperature and diffusivity coefficients were constant. The outer resistance coefficient was considered inconsequentially negligible, and the moisture proportion was assessed as follows (Crank, 1975):

$$MR = \frac{8}{\pi^2} \cdot \exp\left(- \frac{\pi^2 \cdot D_{eff} \cdot t}{4 \cdot L^2} \right) \tag{3}$$

The effective diffusivity was determined by the Arrhenius-like equation (Doymaz, 2012):

$$\ln\left(D_{eff}\right) = \ln(D_0) - \frac{E_a}{R \cdot (T + 273.15)} \tag{4}$$

where: D_0 – the Arrehnius constant (m^2/s), E_a - the activation energy (kJ/mol), T - the air temperature (°C) and R - the universal gas constant (8.314 kJ/mol• K). The Arrhenius diagram was built according to the complementary of the absolute temperature based on the natural logarithm of the evaluated D_{eff}. The acquired curve was assumed as a straight line, while the activation energy was anticipated from the slope of this line.

2.3. ANN modelling

To predict seed weight and moisture during the drying process, oil and FFA content and thousand-seed weight after the 12-month storage period, a multilayer perceptron model (MLP) with three layers (input, hidden and output) was implemented for three ANN for modelling using the drying parameters (such as: variety type, drying temperature, air velocity and drying time, together with initial oil and FFA content and storage type). The model ANN is considered particularly suitable for solving nonlinear problems with constraints (Johnson et al., 2012; Yun et al., 2013; Kleijnen, 2015; Pavlić et al., 2020). The experimental database was normalised before computing ANN to improve the numerical conduct of ANN. In addition, the Broyden-Fletcher-Goldfarb-Shanno (BFGS) algorithm was used to speed up the convergence of the model (Kollo and von Rosen, 2005; Pezo et al., 2013).

The data were randomly divided into training (60%), cross-validation (20 %) and test (20 %) data groups. During the modelling sequence, different ANN architectures were explored. Different numbers of hidden neurons (5−20) and different activation functions in the hidden and output layers were used, and the structure of ANN was reconstructed more than 100,000 times. The arbitrary values of the initial weight and bias coefficients (Ochoa-Martínez and Ayala-Aponte, 2007) were approximated using the ANN training cycle (Doumpos and Zopounidis, 2011). The BFGS algorithm was used to accelerate and stabilise the convergence of the solution for the ANN model (Taylor, 2006).

2.4. Global sensitivity analysis

The formula of Yoon (ANN) (Yoon et al., 2017) was used to investigate the relative influence of hybrid type, drying temperature, air velocity and drying time, and initial oil and FFA content on seed weight and moisture during the drying process, and on oil and FFA content and thousand grain weight after 12 months of storage.

2.5. The accuracy of the model

The created ANN model was inspected utilizing several statistical tests, like the coefficient of determination (r^2), reduced chi-square (χ^2), mean bias error (MBE), root mean square error (RMSE), and mean percentage error (MPE) (Aćimović et al., 2020).

2.6. Multi-objective optimization

The elaborated model ANN was used in the multi-objective

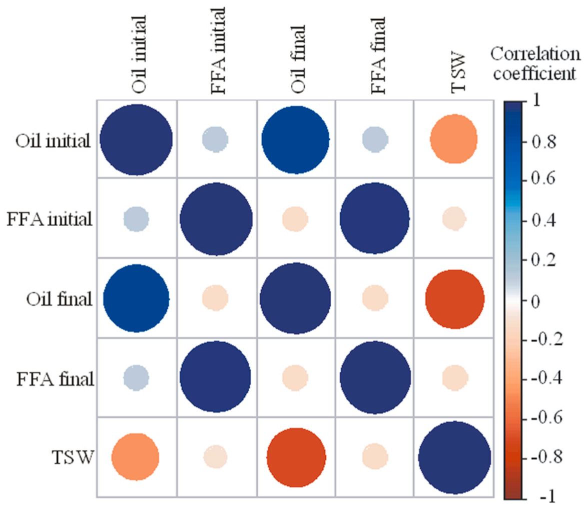

Fig. 2. Colour correlation graph between obtained yields of investigated cultivars of rapeseed.

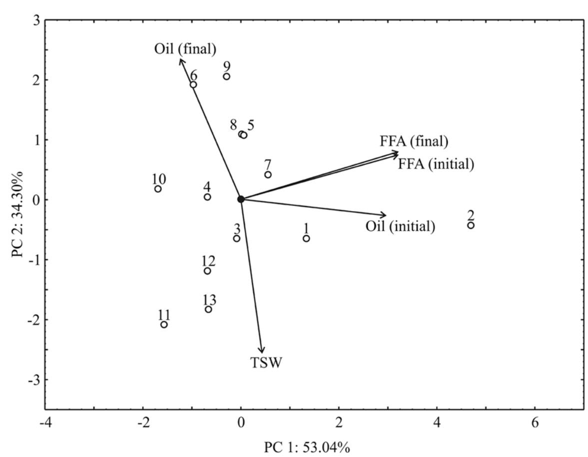

Fig. 3. PCA ordination of variables based on component correlations.

optimisation (MOO) to determine the drying temperature that leads to minimal changes in oil and FFA content and weight of one thousand seeds after twelve (12) months of storage. To solve the MOO (Kojić et al., 2018), a Pareto front with a certain objective function was agreed, and the genetic algorithm (GA) was applied to explore the solutions to the MOO problem (Goldberg, 1989). Populations in the GA were scored according to the distance measure of each point in the current generation (Kojić et al., 2018; Silitonga et al., 2019).

3. Results and discussion

The moisture content, the content of oil and FFA as well as the thousand seed weight before and after drying are presented in Table 2.

Initial oil content was positively correlated with FFA (initial) and FFA (final) at $p \leq 0.05$ level. The correlation between the initial and final FFA content was statistically significant at $p \leq 0.01$ level. The obtained correlations were plotted in Fig. 2 with the function "corrplot" using the method "circle" from the programme R Studio 1.4.1106. The size and colour of the circle depend on the correlation coefficients. If the colour is blue, a positive correlation was obtained, whereas the red colour stands for a negative correlation. Furthermore, the size of the circle is increased with the absolute value of the correlation coefficient.

Before drying, the oil content ranged from 41.2 % to 46.0 % and FFA from 0.6 % to 2.3 % and after drying the oil content ranged from 39.6 % to 44.8% and FFA from 0.9 % to 2.6 %. The weight of thousand-seed varied between 2.8 and 6.3 g.

The mass of thousand-seed of the varieties studied was monitored during storage. From the analysis of rapeseed storage, it can be concluded that the variation in the mass of thousand-seed was greatest for naturally dried seeds and for seeds stored under environmental conditions. This is also logical, because when relative humidity increases, grain moisture also increases and vice versa. For example, Izli et al. (2009) found that increasing the moisture content of rapeseed increased some physical properties, including thousand-seed weight. The same conclusion about the relationship between seed moisture content and thousand-seed weight was drawn by Razavi et al. (2009).

It is noted that the most significant loss of oil and the greatest increase in FFA content occurred during storage of 12 months under ambient conditions, regardless of the temperature of drying prior to seed storage. At the same time, there were no significant differences in losses between storage at 4 °C and 10 °C. At the end of rapeseed storage, an increase in FFA content was observed. The greatest difference FFA content before and after storage of 12 months was found under conditions where air temperature and humidity changed in response to changes in these parameters in the ambient air (storage under ambient air conditions) (Rokosik et al., 2019).

The intensity of the biological and chemical processes that take place in rapeseed depends on the storage conditions. The optimal moisture content of seed intended for long-term storage should be 7 % (w.b). Storage of rapeseed with increased humidity leads to an increase in lipase and microbiological activity which leads to an increase in the content of FFA. The oxidation and degradation of these compounds significantly reduce the value of the oil for extraction (Gawrysiak-Witulska et al., 2018). Sun et al. (2014) stored high and low rapeseed varieties with a moisture content of 8 %, 10 %, 12 %, and 14 % (w.b.) at 10, 20, 30, and 40 °C for 20 weeks and analysed the quality changes of rapeseed during storage. The authors found that the FFA value of rapeseed stored at 10, 20, and 30 °C increased with increasing storage time, but for rapeseed stored at 40 °C, the FFA value increased up to 6 weeks storage and then dropped dramatically. The authors concluded that safe storage moisture is lower in high oil rapeseed than in low oil varieties. As the moisture content of the seeds at the end of drying was about 6 % for all varieties and all treatments, no large increase in FFA content was observed.

The PCA of the experimentally obtained data explained that the first two components accounted for 87.35 % of the total variance (53.04 % and 34.30 %, respectively) in the five variables (yields obtained from the rapeseed varieties studied). Looking at the map of PCA performed on the data, the initial and final contents FFA (which, according to the correlations, accounted for 33.10% and 32.9% of the total variance, respectively) and the initial oil content (28.3 %) have positive values according to the first principal component (Fig. 3). The positive contribution to the calculation of the second principal component was observed for the final oil content (41.3 % of the total variance, based on the correlations), while negative values were observed for the TSW (49.6 %) in the calculation of the second principal component.

After measuring the initial moisture, oil and FFA content, the drying process was continued to a value of 6% of the average moisture. The oil content before drying was between 41.2 % and 46.0 % and FFA between 0.6 % and 2.3 %. After drying, the oil content was between 39.6% and 44.8% and FFA between 0.9 % and 2.6 %. The weight of a thousand-seed was between 2.8 and 6.3 g. It is noted that the greatest loss of oil and the greatest increase in FFA content occurred during storage under ambient conditions regardless of temperature, while there were no significant differences between storage at 4 °C and 10 °C. In addition, the mass of thousand-seed of the hybrids studied was monitored during storage. Based on the study of oilseed rape storage, it can be claimed that the variation in the mass of thousand-seed was greatest for naturally dried seeds and for seeds stored under environmental conditions.

Experimental results of drying process for 13 rapeseed varieties were

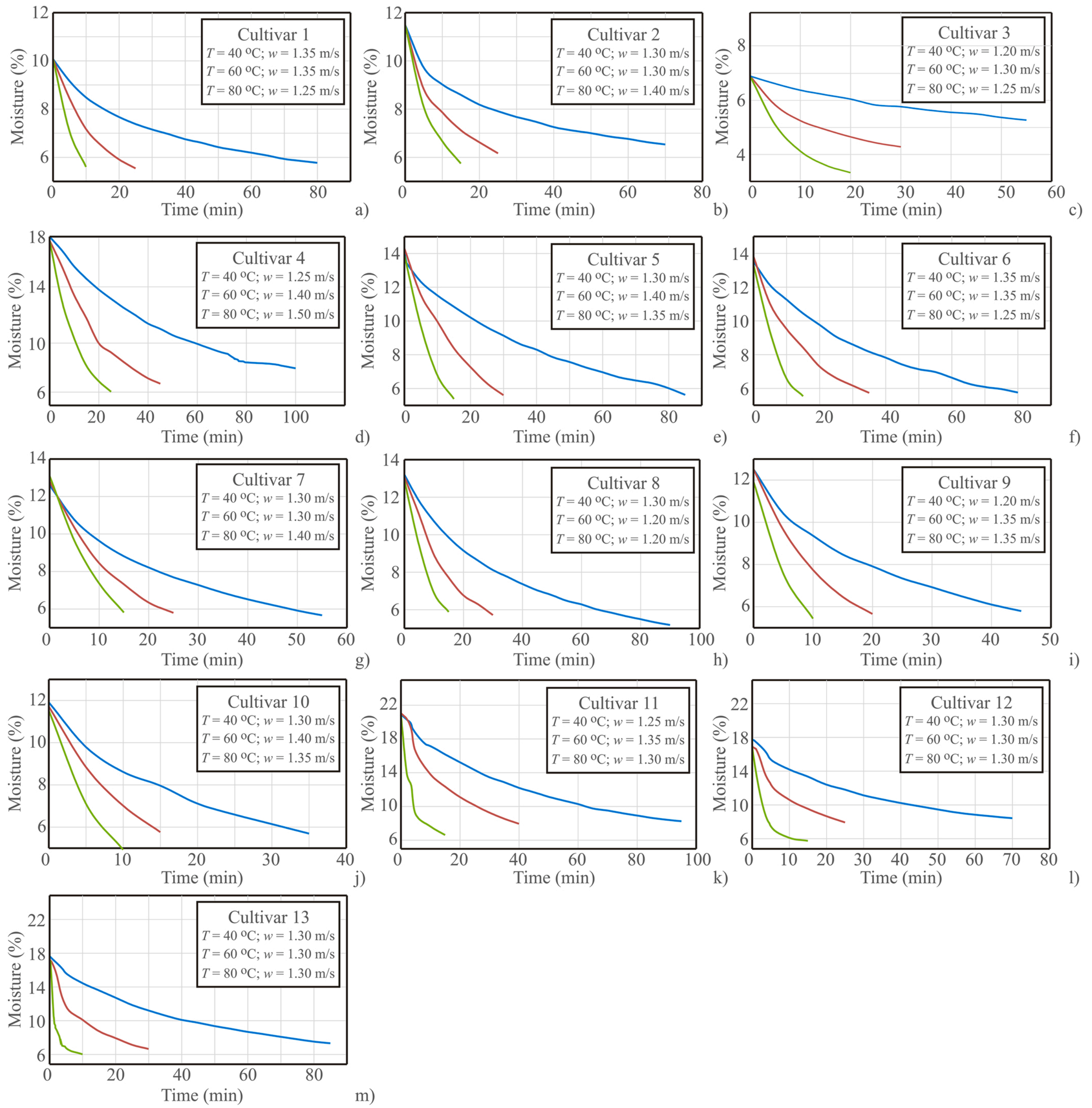

Fig. 4. Experimental results of moisture during the drying process for 13 Croatian rapeseed varieties, according to different temperatures and different air velocities.

presented in Fig. 4, where the moisture of samples was recorded, according to different temperature and different air velocity.

3.1. Determination of the effective moisture diffusivity

The calculated D_{eff} values obtained at specific temperatures are shown in Table 3. The effective diffusivity ranged from: $7.947 \bullet 10^{-10}$ to $1.459 \bullet 10^{-8}$ m^2/s (first drying period) and $4.716 \bullet 10^{-10}$ to $8.611 \bullet 10^{-9}$ m^2/s (second drying period). The evaluated D_{eff} values showed a positive correlation with the increase in temperature. The analogous results for food drying (10^{-12}-10^{-8} m^2/s) are found in the literature (Zogzas et al., 1996), while the effective diffusivities for rapeseed drying were: $1.72 \bullet 10^{-11}$, $2.41 \bullet 10^{-11}$ and $3.31 \bullet 10^{-11}$ m^2/s for 40, 50 and 60°C, respectively (Le Ahn et al., 2008). The activation energy for thirteen rapeseed hybrids was also presented in Table 3. In accordance with the Arrhenius equation, the activation energy was approximated between 17.169 and 42.546 kJ/mol (first drying period) and between 31.261 and 50.474 kJ/mol (second drying period). The determined activation energy data agreed quite well with the literature data, according to which the activation energy for the second drying period was between 28.47 kJ/mol (Chayjan et al., 2011).

The dependence of the moisture content during harvesting and 1000-kernels weight on D_{eff} values throughout the drying process for 13 Croatian rapeseed varieties, according to a different temperature. The diffusion of moisture was more pronounced in case the moisture content was closer to 14 % and if the temperature of drying was more augmented

Table 3
Effective moisture diffusivity (m^2/s) and activation energies (kJ/mol), for thirteen rapeseed hybrids at temperatures between 40 and 80 °C for the first and the second drying periods.

Cultivar	Temperature (°C)	First period D_{eff} ($\times10^{-9}$ m^2/s)	First period E_a (kJ/mol)	Second period D_{eff} ($\times10^{-9}$ m^2/s)	Second period E_a (kJ/mol)
1	40	0.633	50.474	1.743	27.610
	60	2.412		3.445	
	80	5.951		5.951	
2	40	0.675	43.369	2.454	18.204
	60	2.363		3.850	
	80	4.613		5.515	
3	40	0.472	46.174	0.795	42.546
	60	1.522		2.772	
	80	3.672		5.230	
4	40	0.986	36.138	1.530	31.058
	60	2.505		2.966	
	80	4.911		6.117	
5	40	0.983	42.565	1.692	31.964
	60	3.140		3.159	
	80	6.498		7.060	
6	40	1.035	39.618	1.840	30.495
	60	2.483		3.776	
	80	6.053		7.153	
7	40	1.373	31.261	2.726	17.169
	60	3.230		4.224	
	80	5.495		5.847	
8	40	0.984	38.407	2.111	24.751
	60	2.823		3.887	
	80	5.410		6.346	
9	40	1.619	35.828	2.934	22.390
	60	4.031		4.861	
	80	7.957		7.957	
10	40	2.030	32.540	3.282	21.618
	60	4.781		5.195	
	80	8.611		8.611	
11	40	0.978	44.358	4.398	26.328
	60	2.495		7.133	
	80	7.078		14.283	
12	40	1.054	37.916	4.079	28.748
	60	3.172		9.164	
	80	5.654		14.591	
13	40	1.019	41.867	3.303	30.781
	60	3.239		11.311	
	80	6.524		12.737	

(Fig. 5a). The lower or the higher moisture content than 13 % leads to decrease of D_{eff} values. Also, according to results the diffusion rate of moisture was notable lower in the case of the 1000-kernels weight was near to 5 g (Fig. 5b), due to increased active surface for heat exchange (Crank, 1975).

3.2. Artificial neural network model (ANN)

Quite possibly, ANN models are one of the most successful modelling tools in elucidating explicit designing problems, particularly for profoundly interconnected parameter structures where nonlinear connections and relationships occur (Huang et al., 2010). Due to their adaptability, high prescient exactness, high generalisation capacity, and insensitivity toward noisy data, the implementation of ANN is by all accounts a promising approach to modelling complex systems (Du and Sun, 2006). Since a bulk material is a dynamic multiplex environment wherein a large number of nonlinear collaborations occur, the investigation endeavoured to use ANN to foster a predictive model for the storage changes of a bulk material dried at different air temperatures. For this reason, data were used covering a wide range of storage parameters that may occur in agricultural practice during drying at 4 different temperatures and storage under 3 different storage conditions.

The designed optimal neural network models (ANN1, ANN2, and ANN3) presented in Fig. 6 had suitable generalisation properties to

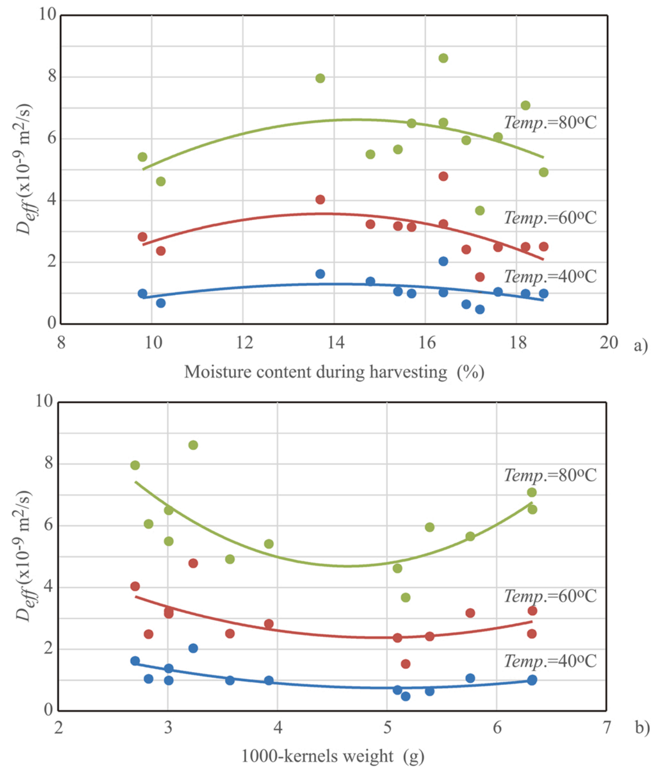

Fig. 5. The influence of a) the moisture content during harvesting and b) 1000-kernels weight on D_{eff} values throughout the drying process for 13 Croatian rapeseed varieties, according to different temperature.

communicate the experimental data. Along these lines, they could be used to foresee the weight and moisture of the seeds during the drying process and the oil and FFA content in a thousand seeds following one year of storage as a function of the drying parameters (hybrid type, drying temperature, airspeed and drying duration, initial oil and FFA content and drying type).

According to the ANN model computation, the optimal number of neurons in the hidden layers for the ANN1, ANN2, and ANN3 models were: 7, 6, and 6 (networks MLP 4–7–2, MLP 7–6–2 and MLP 5–6–1, respectively). Moreover, the obtained models had high r^2 values; during the learning cycle, the r^2 values reached: 0.983, 0.955, and 0.926 for models ANN1, ANN2, and ANN3, individually Table 4.

The elements of the matrices (weighting and bias coefficients) which represented the ANN models, were written in the supplementary tables 1–5.

The constructed ANN models for predicting the output parameters were challenging (with 57, 62 and 43 wtbiases coefficients for ANN1, ANN2, and ANN3, respectively), consistent with the strong nonlinearity of the tested ANN models (Montgomery, 1984; Chattopadhyay and Rangarajan, 2014). Nevertheless, the accuracy of the ANN model could be visually established from the distribution of the experimentally determined values and the ANN calculated values, as shown in the graph in Fig. 7. The calculated values of the ANN models were close to the experimental values in most cases.

The fitting of the ANN model's computation to the experimental data was displayed in Table 5.

The ANN model clearly anticipated the experimentally resolved parameters for a wide range of values. In most cases, the ANN model expected values close to the experimental data (according to the r^2 values). The SOS values calculated with the developed ANN models were similar to the literature values (Kollo and von Rosen, 2005; Doumpos and Zopounidis, 2011). A negligible lack of fit was found for the models, which means that the model predicts the initial parameters satisfactorily

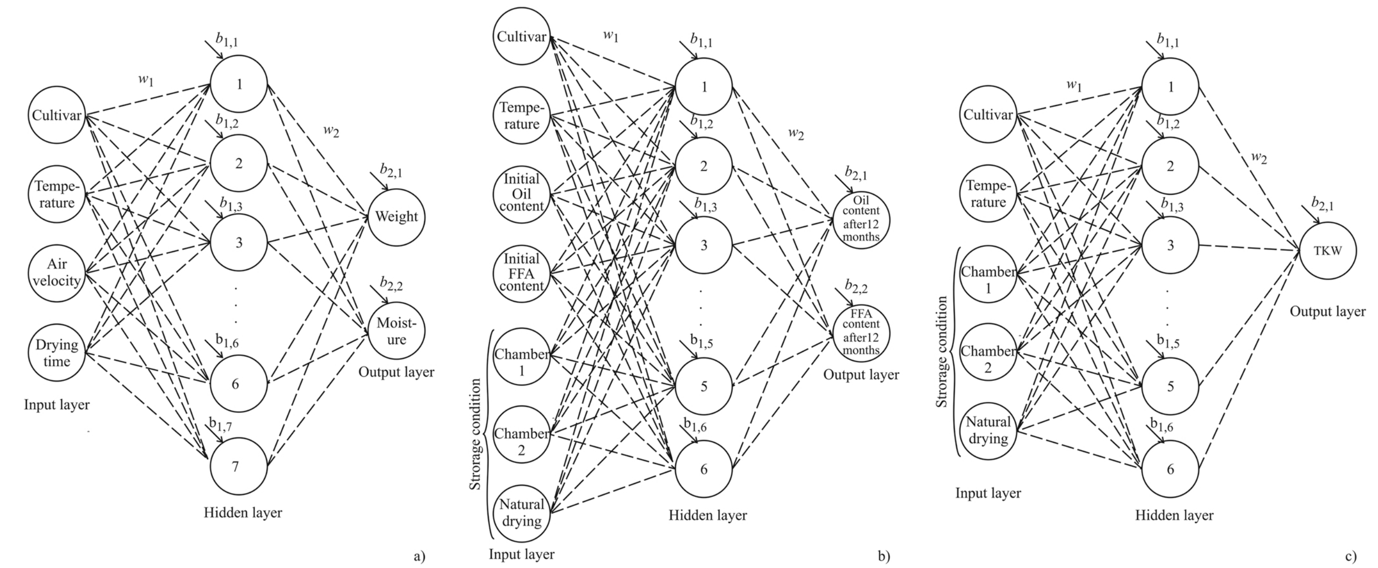

Fig. 6. Schematic presentation of artificial neural network models: a) ANN1, b) ANN2 and c) ANN3. FFA – free fatty acid content, TKW - 1000-kernel weight.

Table 4
ANN model summary (according to performance and errors), for training, testing, and validation cycles.

Network name	Performance			Error			Training algorithm	Error function	Hidden activation	Output activation
	Train.	Test.	Valid.	Train.	Test.	Valid.				
MLP 4–7–2	0.983	0.983	0.984	0.205	0.231	0.293	BFGS 218	SOS	Tanh	Logistic
MLP 7–6–2	0.955	0.958	0.943	0.224	0.267	0.257	BFGS 125	SOS	Logistic	Identity
MLP 5–6–1	0.926	0.910	0.865	0.166	0.185	0.302	BFGS 164	SOS	Tanh	Tanh

*Performance term represents the coefficients of determination, while error terms indicate a lack of fitting for the ANN model

(high r^2 value) and the changes in the predicted data are sufficiently small to account for the experimental values (Erbay and Icier, 2009; Turanyi and Tomlin, 2014).

The discrepancy between the experimental data and the models from ANN was investigated using residual analysis. According to the statistical relationship between the explanatory and output variables, the calculated residuals approached the random errors, which were insignificant. The results of the residual analysis are presented in Table 6. They show that the occurrence of the residuals was random, indicating that the model fits the data well.

The skewness parameters calculate the deviation of the distribution from normal symmetry. The skewness determined was a value close to zero, which means that the distribution of the variables was asymmetrical. The kurtosis parameter expresses the "peakedness" of a variable distribution. The kurtosis was obviously close to zero, indicating a normal distribution.

3.3. Yoon's interpretation method

This chapter investigated the factor's impacts (such as weight and moisture of the seeds) during drying, while the initial oil and FFA amount during drying and the drying temperatures were used to study the oil and FFA content and the thousand seeds after one year of drying. As illustrated in Fig. 8, cultivar type was the most influential parameter in assessing seed weight and moisture with a relative importance of − 47.75% and − 51.79%, individually, while air velocity was influenced by air velocity + 33.38% and − 23.81%, separately. Furthermore, initial oil content and free fatty acid content were the most influential variables for oil content and fatty acid content after one year of storage, while variety was the most crucial variable for ascertaining TKW.

3.4. Multi-objective optimization of the outputs of the ANN

Optimisation of the performance of ANN was carried out by applying the collected data in Eq. 4. One of the main objectives of this study was to maximise seed weight, oil, and FFA content and thousand seed weight and minimise moisture after the drying process in parallel i.e., using ANN1, ANN2, and ANN3 models while varying the input parameters. The required mathematical tasks were solved using the MOO calculation in Matlab. The method MOO included maximisation of seed weight and minimisation of moisture content for the ANN1 model, maximisation of oil and FFA content for the ANN2 model and furthermore maximisation of thousand seed weight for the ANN3 model. The necessary constraints for the optimisation were used within the experimental range of the variables. The number of generations was 739 for 392 and 312 for the ANN1, ANN2 and ANN3 models, respectively. The population proportion was set to 100 for all three models for each input variable. After the Pareto front, the number of points was 39, 21 and 17 for models ANN1, ANN2 and ANN3, respectively. The calculated maximum values for seed weight, oil content, FFA and thousand-seed weight during the drying process were: 500.0 g; 46.0 g; 3.7 g and 8.3 g, respectively, and the minimum moisture content was 3.3%. The oil and FFA contents were 42.2% and 1.38%, respectively, while the weight of thousand-seed reached 5.1 g. The optimum result was obtained at a drying temperature of 80ºC for rapeseed hybrid 3 and an airspeed of 1.25 m/s for 20 min. The drying process was carried out in a Type I procedure.

The study has shown that ANN can be effectively used as a modelling method for predicting the drying and storage of rapeseed. Furthermore, the satisfactory accuracy and precision of the developed ANN models indicate that they can be applied in the postharvest management systems for rapeseed as a supporting tool and then used as a crucial supporting tool in controlling the postharvest conservation process.

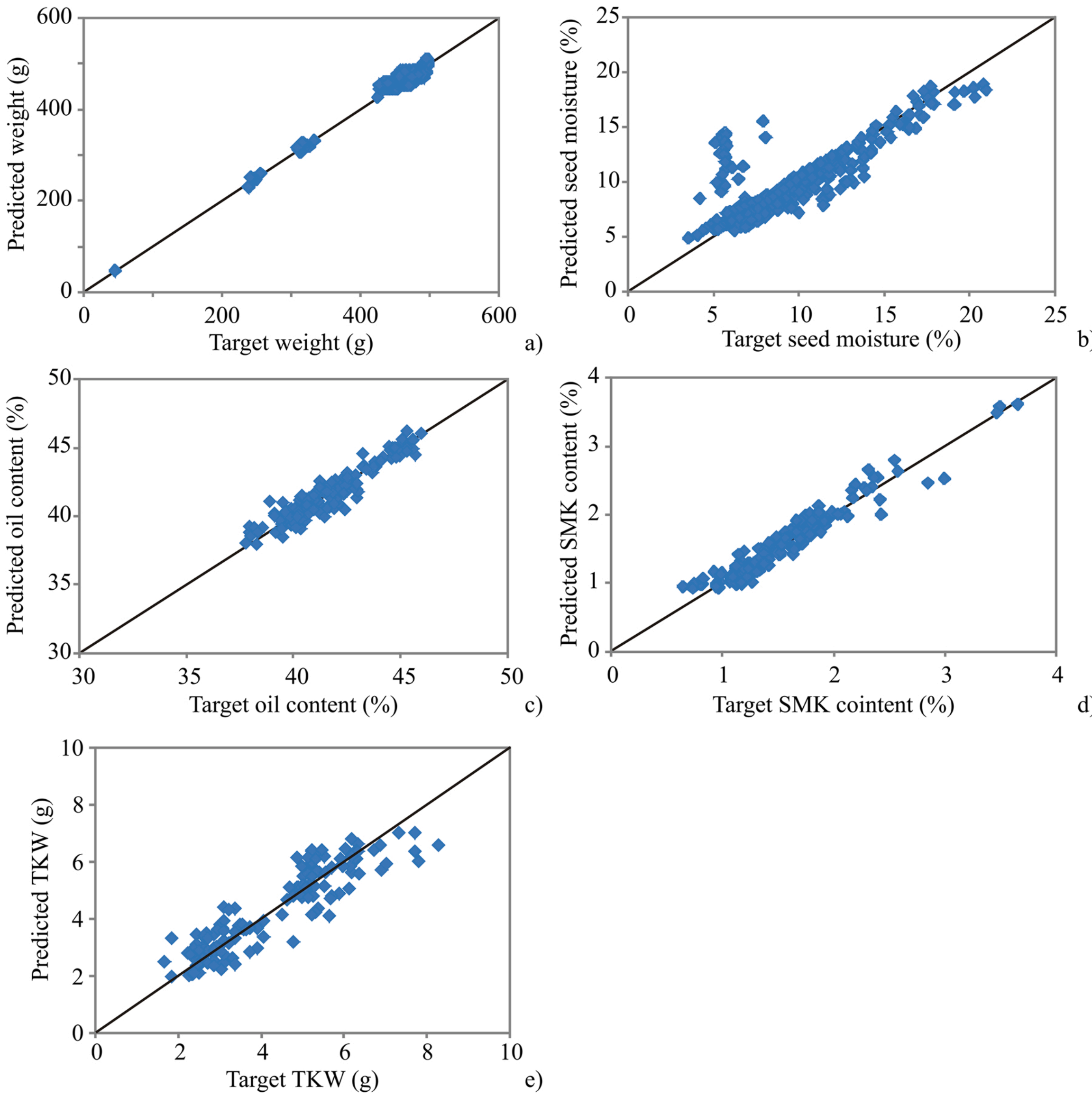

Fig. 7. Values measured experimentally and calculated with the model ANN for the weight and moisture of the seed during the drying process as well as for the oil and FFA content and the thousand seeds weight after one year of storage.

Table 5
The "goodness of fit" tests for the developed ANN model.

Output variable	χ^2	RMSE	MBE	MPE	SSE	AARD	r^2
Weight	1.0E+ 02	1.0E+ 01	5.3E-03	1.763	3.6E+ 04	7.3E+ 03	0.965
Moisture	4.5E-01	6.7E-01	5.2E-02	5.744	1.6E+ 02	1.8E+ 02	0.965
Oil	4.5E-01	6.7E-01	7.2E-02	3.294	1.6E+ 02	2.1E+ 02	0.998
FFA	2.6E-01	5.1E-01	5.2E-02	5.877	9.3E+ 01	1.3E+ 02	0.990
TKW	4.2E-01	6.5E-01	5.7E-02	7.733	1.5E+ 02	3.7E+ 02	0.976

χ^2– reduced chi-square, RMSE - root mean square error, MBE – mean bias error, MPE – mean percentage error, SSE – sum of squared errors, AARD – absolute average relative deviation, r^2– coefficient of determination

Table 6
The "goodness of fit" tests for the developed ANN model.

Output variable	Skew	Kurt	Mean	StDev	Var
Weight	-0.229	-0.195	5.3E-03	1.0E+ 01	3.6E+ 02
Moisture	0.164	1.837	5.2E-02	6.7E-01	3.6E+ 02
Oil	0.250	1.348	7.2E-02	6.7E-01	3.6E+ 02
FFA	0.847	5.630	5.2E-02	5.1E-01	3.6E+ 02
TKW	0.453	1.551	5.7E-02	6.5E-01	3.6E+ 02

4. Conclusion

The obtained effective diffusivity coefficients reached values between $7.947 \bullet 10^{-10}$ and $1.459 \bullet 10^{-8}$ m^2/s for the first drying period and values between $4.716 \bullet 10^{-10}$ and $8.611 \bullet 10^{-9}$ m^2/s (for the second drying period). Furthermore, the evaluated D_{eff} values showed a positive correlation with the increase in temperature.

Furthermore, the application of the developed ANN showed to be a promising, scientifically confirmed approach to predict changes in

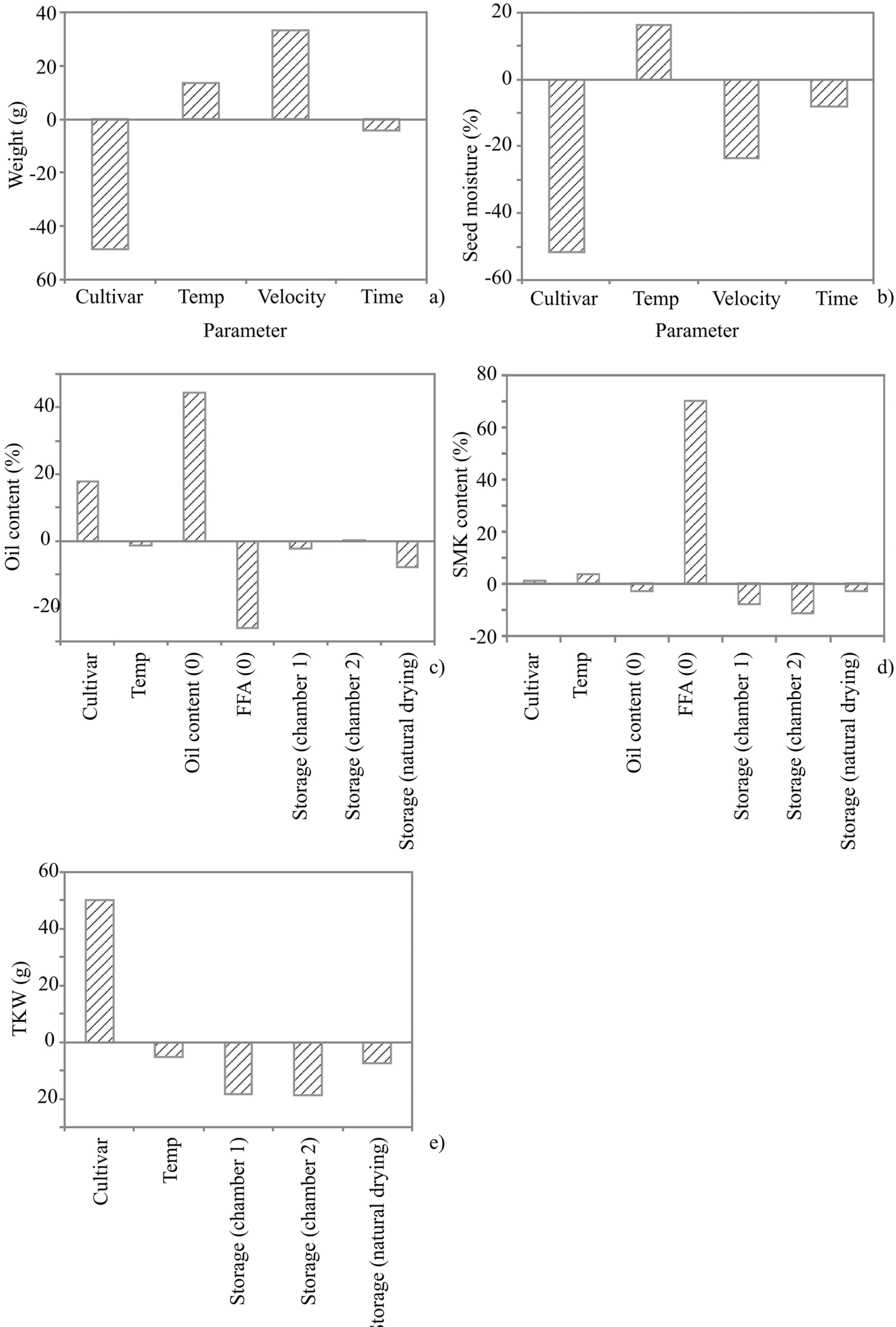

Fig. 8. The relative importance of the input variables on outputs, determined using the Yoon interpretation method.

Table A1
Elements of matrix W_1 and vector B_1 (presented in the bias row).

	1	2	3	4	5	6	7	8
Cultivar	595.903	184.146	0.025	-327.629	-92.830	-357.857	-117.410	-14.313
Temp	-286.801	140.053	-0.167	263.453	233.745	288.099	279.322	-5.011
Air velocity	-604.418	209.295	-0.094	313.353	117.181	343.794	35.660	-16.087
Time	-0.052	90.045	98.825	-0.065	-82.717	-0.075	-55.352	-55.750
Bias	-400.541	200.218	3.366	57.148	-65.020	62.179	-37.386	27.913

Table A2
Elements of matrix W_2 and vector B_2 (presented in the bias column).

	1	2	3	4	5	6	7	8	Bias
Weight	-182.680	-103.656	-13.831	115.070	0.538	-115.066	-0.547	0.028	-64.264
Moisture	33.244	32.712	-16.795	0.775	-19.565	-0.398	5.369		9.233

Table A3
Elements of matrix W_1 and vector B_1 (presented in the bias row).

	1	2	3	4	5	6
Cultivar	-0.013	-5.102	-2.316	-5.620	-6.102	-0.325
Temp	-1.576	-0.105	2.843	0.330	-0.221	4.962
Oil	1.207	0.704	0.715	1.718	1.348	0.770
FFA	0.980	-0.759	-3.947	-0.809	-2.010	2.060
Storage (type 1)	-0.187	0.474	0.411	0.405	0.628	-0.969
Storage (type2)	-0.255	0.069	0.928	0.043	0.290	0.884
Storage (natural)	-0.283	0.494	-0.737	0.851	0.709	-1.909
Bias	-0.850	0.984	0.672	1.383	1.572	-2.081

Table A4
Elements of matrix W_2 and vector B_2 (presented in the bias column).

	1	2	3	4	5	6	Bias
Oil	1.390	-1.511	0.944	1.565	-0.710	-0.281	-0.284
FFA	0.287	2.264	-0.231	0.830	-2.587	0.144	0.098

Table A5
Elements of matrix W_1 and vector B_1 (presented in the bias row).

	1	2	3	4	5	6
Cultivar	-10.429	9.489	13.686	-2.734	0.402	-15.158
Temp	-0.024	0.217	1.527	-0.551	-0.157	-0.720
Storage (type 1)	1.921	-0.702	-2.947	0.468	-1.076	3.142
Storage (type2)	1.948	-0.651	-2.798	0.493	-1.040	3.059
Storage (natural)	1.931	-0.474	-2.812	0.628	3.669	2.977
Bias	5.823	-1.759	-8.498	1.773	1.486	8.935

Table A6
Elements of matrix W_2 and vector B_2 (presented in the bias column).

	1	2	3	4	5	6	Bias
TKW	1.429	-0.404	-2.146	-0.998	0.215	-3.308	0.859

rapeseed during drying and storage and could therefore be used as decision support for technological postharvest systems. This is confirmed by the results of this study, in which ANN was used to predict the storage characteristics of 13 rapeseed varieties grown in Croatia that may occur in postharvest practices.

The created ANN model of seed properties such as oil and FFA content and thousand-seed weight during the drying process could be predicted depending on the drying parameters (such as variety, drying temperature, air and drying time as well as initial oil and FFA content and storage type). The ANN model was suitable for predicting the initial variables (the r^2 values during the training cycle for these variables seed weight and moisture, oil, and FFA content and thousand-seed weight during the drying process were: 0.965; 0.965; 0.998; 0.990, and 0.976, respectively).

The developed ANN model could be a useful tool to be applied in the field of postharvest technology, especially to improve the current systems for drying and storing rapeseed. The introduction of ANN as a predictive tool in postharvest systems for rapeseed will make it possible to estimate losses during rapeseed processing and thereby take appropriate measures to maintain seed quality.

CRediT authorship contribution statement

Neven Voća: Conceptualization, Methodology, Investigation; Writing – original draft preparation, Writing – review & editing. **Lato Pezo**: Conceptualization, Data curation, Visualization, Writing – original draft preparation, Writing – review & editing. **Željko Jukić**: Methodology, Investigation, Writing – original draft preparation, Writing – review & editing, Validation, **Biljana Lončar**: Data curation, Visualization, Writing – original draft preparation, Writing – review & editing. **Danijela Šuput**: Data curation, Visualization, Writing – original draft preparation, Writing – review & editing; **Tajana Krička**: Writing – review & editing, Supervision, Validation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Appendix

See Tables A1-A6 in appendix section.

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