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Mathematical Modeling and Optimization of Ultrasonic Pre-Treatment for Drying of Pumpkin (*Cucurbita moschata*)

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Abstract: Innovations in food drying processes are usually aimed at reducing drying time and improving the overall properties of dried products. These are important issues from an economic and environmental point of view and can contribute to the sustainability of the whole process. In this study, the effects of ultrasonic treatment on the drying kinetics of pumpkin pulp are investigated, and mathematical models to predict the drying kinetics are analyzed and optimized. The results show that ultrasonic pretreatment significantly reduces drying time from 451 to 268 min, with optimal processing parameters at 90% of the maximum ultrasonic power and a processing time of 45 min. The total color change of the samples was the lowest at the obtained optimal processing parameters. Based on the values (RMSE and R^2) of the investigated mathematical drying models, it was found that the Weibull model is the best fit for the experimental data and is considered suitable for the drying kinetics of ultrasonically pretreated pumpkin samples. In this study, an artificial neural network with 15 neurons in hidden layers was also used to model the drying process in combination with ultrasound pretreatment. The network had a performance of 0.999987 and the mean square error was 8.03×10^{-5}, showing how artificial neural networks can successfully predict the effects of all tested process variables on the drying time/moisture ratio.

Keywords: pumpkin; drying; ultrasonics; mathematical models

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1. Introduction

The pumpkin belongs to the wide family of cucurbits (Cucurbitaceae), and the three most common varieties of pumpkin are called *Cucurbita maxima*, *Cucurbita moschata*, and *Cucurbita pepo* [1,2]. Europe is the second-largest producer with approximately 4.9 million tons of pumpkin, which is about 17.5% of world production [3]. However, since pumpkin is mainly used for seeds, decorative purposes, and as animal feed, large amounts of waste and by-products are generated during industrial processing. In general, seeds represent 10% of the total weight, whereas about 90% of pulp and peel are discarded, having previously been considered as waste [4]. The pulp and peel contain many functional compounds such as polyphenols, carotenoids (mainly β-carotene), vitamin C, low-energy sugars, and a large amount of dietary fiber [5]. Pumpkin, as a cultivar that is widespread throughout the world, is recognized as one of the three medicinal plants beneficial for diabetes [6]. The chemical and biochemical composition of pumpkin has been extensively researched and it has been proven that pumpkin is a rich source of vitamins and minerals [7], essential nutrients, phenols, flavonoids, and carotenoids [8], and antioxidants [9]. Thus, pumpkin has beneficial effects on human health [8], reduces the risk of neurodegenerative, cardiovascular, and cancer diseases [10] and prevents osteoporosis and hypertension [11,12].

Drying is the most commonly used method for food preservation and extending its shelf life [13,14]. Due to rising energy prices, food drying is an energy-intensive process that consumes up to 15% of energy in all food industries combined [15]. There is a growing interest in the use of ultrasound in co- or pre-processing for drying. The use of ultrasound in the treatment of food raw materials, wastes and by-products shows promising results for versatile applications, both in primary research and in industry [16–20].

In general, sonication/ultrasound represents a sound that is inaudible to humans because the ultrasound frequency starts at 18 kHz, which is inaudible to humans. Ultrasound with such power is used in a wide variety of applications, such as extraction, emulsification, homogenization, sieving, sedimentation, micronization, pasteurization, cell disruption, drug delivery, sterilization, wastewater treatment, and in general food processing [21–24].

Low-frequency ultrasound (18–100 kHz), with low to high intensities, expressed by the diameter of the probe (5–1000 W/cm^{-2}), is most commonly used for treatments. Among these ultrasound parameters, the propagation of sound waves showed great potential [25]. When the research aims at the gentle propagation of ultrasound waves considering the applied intensities, ultrasound baths (indirect treatment) are used as devices, while on the other hand, when high intensities are required, devices with directly immersed probes (direct treatments) are used [26].

Figure 1 shows the most common ultrasonic devices used for food processing.

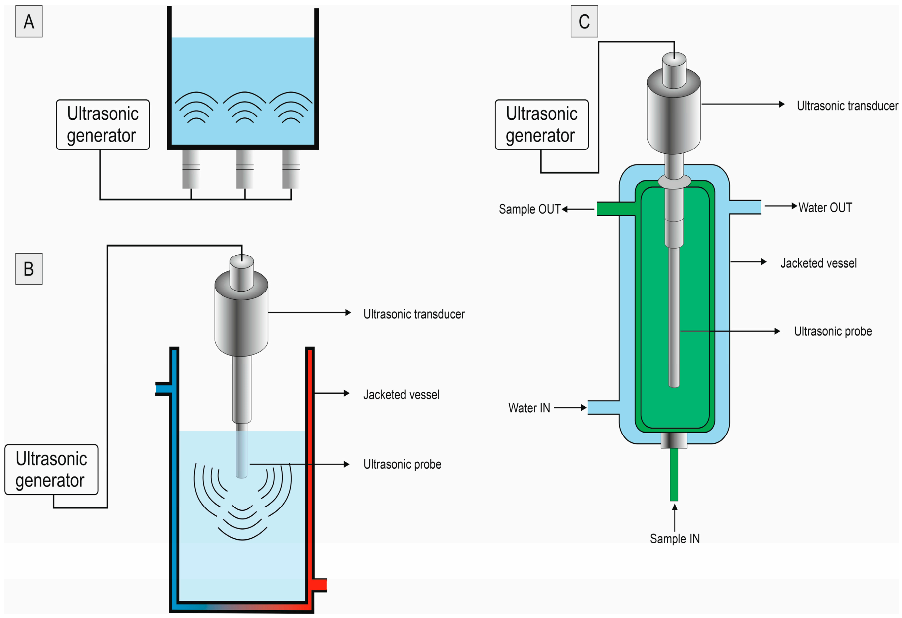

Figure 1. Most common ultrasonic setups: (**A**) bath with probes mounted bellow: (**B**) directly immersed probe: (**C**) continuous flow sonication with cooling.

Each of the presented ultrasound devices is equipped with the following four basic elements: An ultrasound generator, an ultrasound transducer, a probe or probes depending on the system and a treatment chamber as well. The main mechanism of ultrasonic work in the liquid medium is based on the generation and implosion of gas cavitation bubbles in the treated liquid medium. Cavitation bubbles are created in the vicinity of the liquid treated by the ultrasonic waves, which are subjected to rapid and alternating pressure with high amplitude. In the physics of sound, there are negative and positive pressure cycles, i.e., during the negative half of this phenomenon, the treated sample is stretched, while in the positive half it is compressed. The resulting microbubbles vary in size during the negative and positive pressure cycles until the final phenomenon, implosion, occurs.

When the bubbles implode, they release enormous amounts of energy in the form of high pressure (up to 100 MPa) and high temperatures (up to 5000 K) [27]. However, the release of all the accumulated energy results in local pressure and temperature changes that dissipate in the liquid in the chamber. This local energy propagates into the liquid environment and causes structural, chemical, and physical changes in the immersed sample [28]. In addition, ultrasound has the advantage of non-thermal technology and thus has a positive impact on the environment.

Ultrasound as a pretreatment in food drying is gaining increasing attention, as it does not only shorten the drying time, but also helps to reduce energy consumption [29,30]. The mechanical and thermal effects of ultrasonic cavitation can shorten drying time by altering or destroying the cellular matrix. It can also contribute to the removal of the wax coating [31]. Both effects lead to an improvement in the mass and heat transfer with each further drying process (vacuum, microwave, conventional, etc.) [32–34]. The drying process usually has two or three periods. The first is a period in which the drying rate is constant (so that the drying curve is practically linear), followed by a period of decreasing drying rate. During the second period, the water evaporates from the surface of the material, while the water inside the material diffuses to the surface through the pores of the cell matrix. Ultrasonic treatment increases the size and number of pores, thus accelerating mass transfer and shortening the drying time. Mathematical models have been used to describe the kinetics of mass transfer in convective drying processes as a function of drying conditions, such as temperature or pressure. Modeling the drying process can help improve drying efficiency, shorten drying time, and thus improve time and energy efficiency as well as the sustainability of the drying process. Modeling is a much simpler and faster method for predicting optimal process parameters and conditions to achieve desired outcomes, such as product quality. It is also a tool to design and size dryers for specific requirements. Many authors have already analyzed and improved mathematical models for drying various food products such as apples, kiwi and other fruits [35–37]. Some limitations and problems with the drying models are that most models are based on a two-step procedure. In the first step, the best model was calculated based on the dependence between water content and drying time, and various errors such as R^2 or χ^2 were obtained for each model. The second step involves the calculation of model constants and coefficients as a function of process parameters such as drying temperature, air velocity, etc. This approach leads to the continuous development of minimally-modified models that excellently describe the drying curves for a specific drying process. This is due to the obtained coefficients, which have no physical meaning or relation to physical processes such as mass and heat transfer. Therefore, such coefficients cannot be correlated with process parameters. Efforts have been made to create more general empirical mathematical models, with some success. However, relatively new modeling techniques such as the use of artificial neural networks have much greater potential to capture a larger number of parameters. Such heuristic models may be better suited to complex and nonlinear processes such as drying combined with ultrasonic (pre)treatment.

The main objective of this study is to select and develop mathematical models for the drying of pumpkin and to optimize the parameters of ultrasonic drying to minimize the drying time. Based on the results, standard empirical models will be compared to the obtained artificial neural networks.

2. Materials and Methods

2.1. Plant Materials

Fresh pumpkin fruits (*Cucurbita moschata*) that were uniform in size and undamaged were selected in late August (about 110 days after ripening) and purchased at the local market. Because pumpkins can vary significantly in size and other physical characteristics, those selected from the obtained batch were measured. The pumpkins were pear-shaped and elongated, with weights of 2.41 ± 0.35 kg and lengths of 30.72 ± 2.48 cm.

Fruits were transported to the laboratory, washed with tap water, and stored in the refrigerator at 4 ± 1 °C until further use. Before starting the experiment, the fruits were removed from the refrigerator and stored at room temperature (23 ± 1 °C) for about 45 min to acclimate them to room temperature [38,39]. The fruits were then washed, peeled, and cut in half lengthwise with a stainless-steel knife. The seeds and fibrous strands were then separated from the pulp. The pulp was cut into uniform pieces 4–5 mm thick and 15 mm long using a mechanical slicer.

The initial moisture content of the fresh pumpkin was determined by oven drying at 105 °C for 24 h using an electric conduction oven (VO200 PM200 Memmert GmbH, Büchenbach, Germany), as described in AOAC [40]. Three repeated measurements were performed.

2.2. Ultrasound Pretreatment

The prepared pumpkin samples were immersed in an ultrasonic bath filled with 7 L of distilled water (Elmasonic P 300 H, Elma–Hans Schmidbauer Gmbh & Co., Singen, Germany). Sonication was performed at a constant frequency of 37 kHz for amplitudes at 30, 60, and 90%. The processing times were 30, 45, and 60 min. The container with the sample was placed in the same position and the water level in the bath tank was kept at a height of 270 mm [41]. During the treatment, stainless steel meshes were placed on the samples to reduce movement.

2.3. Drying Experiment

Before drying, the water content of each pumpkin was measured using an infrared dryer (LJ16, Mettler-Toledo, Leicester, UK). The average water content of the pumpkin pulp was 92.10 ± 2.18%.

Samples were dried in a VO200 PM200 conduction vacuum dryer (Memmert GmbH + Co. KG, Schwabach, Germany) at a temperature of 60 °C [42] and atmospheric pressure of 1000 mBar. The 360 g of samples was divided into two batches and dried on two stainless-steel shelves (180 g each). Stainless-steel meshes were placed on the samples for better heat transfer and a larger contact surface. Water loss during drying was measured every 10 min using a laboratory balance (Mettler Toledo ME1002TE, Columbus, OH, USA). Drying was carried out until a constant mass was reached.

2.4. Color Measurement

The color of the fresh and dried samples was determined using a colorimeter (Konica Minolta CM-3500d, Tokyo, Japan). The total color change (ΔE) was the parameter used for the overall color difference evaluation between a dried and a fresh sample. Based on the referent sample (fresh pumpkin), ΔE was calculated based on the following equation:

$$\Delta E = \sqrt{\left(L_{ref} - L\right)^2 + \left(a_{ref} - a\right)^2 + \left(b_{ref} - b\right)^2}$$

where L indicates lightness, a is the redness, and b is the yellowness; L_{ref}, a_{ref} and b_{ref} are values for the referent sample; and L, a and b are values of the investigated samples.

2.5. Mathematical Modeling

Mathematical models were selected from the already established simple and more complex models used for the prediction of drying kinetics based on the obtained data, which are presented in Table 1.

Table 1. Mathematical models for convection drying.

Model	Equation	References
Page	$MR = \exp(-kt^n)$	[35,36]
Modified Page	$MR = \exp(-(kt)^n))$	
Weibull	$MR = \exp((-t/\alpha)^\beta)$	
Modified two-term	MR = a*exp(−k*t) + (1 − a)*exp (−k*a*t)	

MR—moisture ratio; a, k—coefficients, n—drying exponent.

Two artificial neural networks (ANN) were trained in Statistica software based on the drying data obtained. As in the modeling, the output variable for both ANNs was the moisture ratio (MR). The input variable for the first ANN (further labeled as ANN-1) was based on the optimal parameters for the process variables based on the statistical analysis. For the second ANN (labeled as ANN-2), data obtained for all process variables (process time 30, 45 and 90 min; amplitude 30, 60 and 90%) were used. Two-thirds of the data was used for training and one-third was used for the validation of the model.

2.6. Data Analysis

All analyses were performed using Statistica 13 software (Tibco Statistica 13.3.0). Values were compared using mean comparisons, ANOVA analysis, and Tukey HSD post hoc test to determine significance. The models presented in Table 1 were fitted using non-linear estimation regression analysis based on the Levenberg–Marquardt method. Evaluation of fit of the selected thin layer drying models was based on the coefficient of determination (R^2) and root mean square error (RMSE). The highest R^2 (closer to unity) value and a low RMSE (closer to zero) value were the primary criteria for the selection of the best model [37].

3. Results and Discussion

3.1. Modeling

One way to differentiate mathematical models for predicting drying kinetics can be based on their perceived complexity. Most basic models, such as that of Page of Lewis, use only one parameter, while more complex models, such as that of Midilli, may have three or more parameters. This could significantly improve fitting to experimental drying data, especially for not-so-standard drying curves compared to those usually observed in the conventional thin-layer drying of fruits and vegetables. In our drying experiments with ultrasonic pretreatment, curves such as sigmoidal curve I and II were obtained. However, due to the very large number of models in the literature and the often insignificant differences in results and errors, it is inefficient and unnecessary to test them all. In order to avoid random selection of the appropriate models and to reduce the time required for the analysis, the selection of the models was based on a review of thin-layer drying models [43,44]. For the purpose of screening and to reduce the number of models presented, the Lewis model was tested; however, we did not observe an adequate fit to the experimental data obtained for the ultrasonically processed samples, with the highest R^2 = 0.817 (at 30%, 30 min). Some other commonly used models such as the Wang and Singh, Geometric, and Singh model were also discarded as these also were not optimal for the drying curves. Therefore, among the simpler models, only the Page and the modified Page model and among the more complex models, the modified two-term model and the Weibull model were selected for modeling.

The calculated coefficients of determination and RMSE values are shown in Tables 1–4. It is evident that despite having only one coefficient, the goodness of fit for untreated samples was best for Page's model with an R^2 of 0.9995.

While the Page model still shows a good fit for the ultrasound pretreatment of the drying process, increasing the ultrasound amplitude leads to a decrease in the coefficient of determination and an increase in the RMSE. The Weibull model showed the best fit for ultrasound treatment with a minimal R^2 = 0.9906 for 60% of the amplitude and maximal R^2 = 0.9991 at 40% of maximal amplitude, which is consistent with other research papers [37–39,45]. The modified two-term model with three variables, which was expected to fit the experimental data best, demonstrated a good enough fit to be used, but the much simpler Page and modified Page models were better. The goodness of fit of all the models tested is in direct correlation to the ultrasound amplitude, since an increase in amplitude leads to a decrease in R^2 values. It is also directly related to the duration of ultrasound treatment, with minimal R^2 obtained for samples with a treatment duration of 60 min. All models show a minimal but visible underestimation of the moisture ratio in the constant rate period and a slight overestimation in the falling rate period, as can be seen in Figure 2 for untreated and treated samples.

Table 2. Coefficient of determination and RMSE of selected mathematical models—30 min treatment.

Model	US Power/%	R^2	RMSE
Page	0	0.9995	0.0010
	30	0.9623	0.0056
	60	0.9784	0.0051
	90	0.9442	0.0078
Modified Page	0	0.9990	0.0015
	30	0.9624	0.0062
	60	0.9790	0.0060
	90	0.9457	0.0083
Weibull	0	0.9990	0.0006
	30	0.9917	0.0011
	60	0.9974	0.0020
	90	0.9940	0.0019
Modified two-term	0	0.9997	0.0018
	30	0.9602	0.0035
	60	0.9762	0.0029
	90	0.9418	0.0054

Table 3. Coefficient of determination and RMSE of selected mathematical models—45 min treatment.

Model	US Power/%	R^2	RMSE
Page	30	0.9981	0.0018
	60	0.9675	0.0044
	90	0.9334	0.0178
Modified Page	30	0.9957	0.0014
	60	0.9652	0.0033
	90	0.9314	0.0081
Weibull	30	0.9991	0.0015
	60	0.9933	0.0025
	90	0.9978	0.0042
Modified two-term	30	0.9929	0.0030
	60	0.9635	0.0187
	90	0.9373	0.0231

Table 4. Coefficient of determination and RMSE of selected mathematical models—60 min treatment.

Model	US Power/%	R^2	RMSE
Page	30	0.9801	0.0029
	60	0.9860	0.0043
	90	0.9421	0.0082
Modified Page	30	0.9759	0.0033
	60	0.9814	0.0027
	90	0.9373	0.0195
Weibull	30	0.9952	0.0019
	60	0.9906	0.0014
	90	0.9968	0.0034
Modified two-term	30	0.9747	0.0034
	60	0.9816	0.0027
	90	0.9288	0.0109

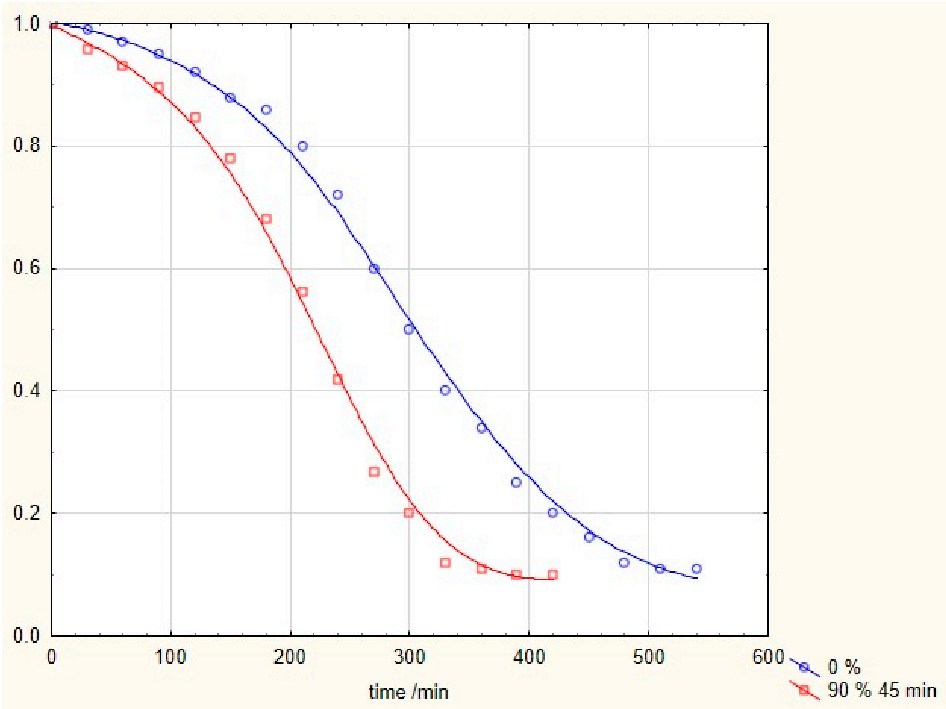

Figure 2. Relationship between the moisture ratio and drying time without US and using optimal US processing time, with Weibull model.

3.2. Optimal Pre-Treatment Parameters

The final dried products are shown in Figure 3. The change in color of the samples was statistically significant (from $\Delta E = 3.95 \pm 0.98$ for samples treated at 30% for 30 min to lowest color change $\Delta E = 3.35 \pm 0.52$ for samples treated at 90% for 60 min (Table 5). As the difference in the color of the samples treated during 30 and 60 min treatments at 90% amplitude was not statistically significant, a shorter processing time seems to be more suitable. However, compared to the duration of ultrasound pre-treatment, it can be observed that the dried pumpkin was lighter (L*) after 45 and 60 min than after 30 min of sonication. These results could be explained by the fact that the ultrasound treatment leads to cavitation, which causes a structural change in the enzymes responsible for the undesirable brown color (polyphenol oxidases), and consequently inhibits the browning of the pumpkin. The relation of ultrasonic parameters to the drying time is presented in Table 6. As expected, the drying time without ultrasound treatment was the longest (452 min), and it can be observed that each ultrasound parameter tested significantly reduced the drying time. A possible consequence of the mechanical and thermal effects of cavitation on all treated samples is the change of the pumpkin matrix and consequently the enlargement of the pores. It also leads to a decrease in the adhesion of water molecules bound to the cell walls [38]. Numerous studies have reported that cavitation phenomena cause a change in the structure of the product and in this way facilitate the faster removal of moisture from the product. Liu et al. reported how the microstructure of purple-fleshed sweet potatoes changed after ultrasonic treatment, showing more microchannels and expanded intercellular spaces. Chao et al. also confirmed that ultrasound pretreatments significantly accelerated the drying rate of seed-used pumpkin due to cell-structure destruction [46–49]. A larger number of larger pores also leads to an increased mass transfer of water during drying and shortens the drying time.

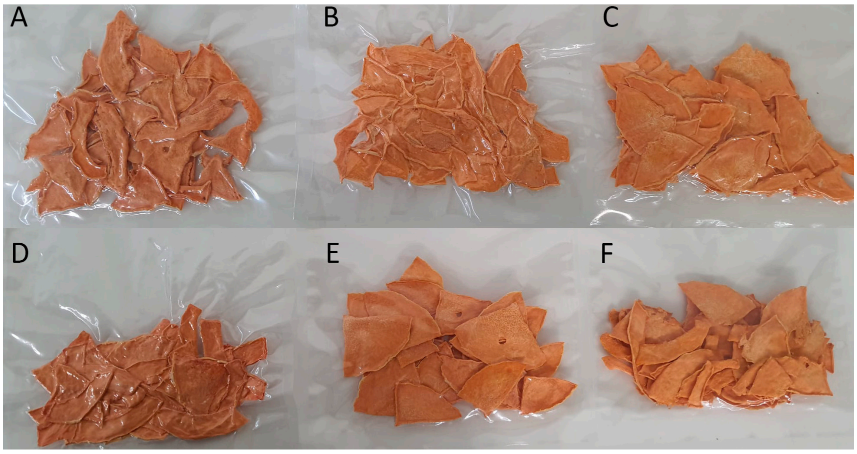

Figure 3. Dried pumpkin slices. (**A–C**)—30% amplitude, 30, 45 and 60 min processing time; (**D–F**)—90% amplitude, 30, 45 and 60 min, respectively.

Table 5. Relationship between ultrasonic parameters and color change in samples.

US Power/%	Sonication Time/min	Total Color Change/Δ*E*
30	30	3.95 ± 0.98a
30	45	4.11 ± 1.19b
30	60	3.87 ± 0.55a
60	30	4.26 ± 0.12b
60	45	4.18 ± 1.07b
60	60	4.17 ± 1.12b
90	30	3.63 ± 0.58a
90	45	3.40 ± 0.57c
90	60	3.35 ± 0.52c

abc—different superscript letters within columns are significantly different ($p < 0.05$).

Table 6. Relationship between the ultrasonic parameters and drying time.

US Power/%	Sonication Time/min	Drying Time/min
0	0	451.67 ± 7.64a
30	30	361.67 ± 2.89b
30	45	356.66 ± 2.89c
30	60	351.67 ± 2.89c
60	30	358.33 ± 2.89b
60	45	346.67 ± 2.89c
60	60	343.33 ± 2.89c
90	30	349.00 ± 5.00d
90	45	322.33 ± 2.89e
90	60	320.67 ± 2.89e

abc—different superscript letters within columns are significantly different ($p < 0.05$).

The statistical analysis of the studied ultrasound parameters, shown in Figure 2, revealed that the shortest drying time was obtained at a maximum ultrasound amplitude of 90% for 45 min. With these parameters, the water content in the samples after drying was 8%, which is within the expected range. Further increasing the processing time had no statistically significant effect on the drying time nor caused an additional reduction in water content.

One factor that had an impact on the longer drying times that correlated with the longer processing times was the initial greater mass of water after processing, and consequently, before drying. This may be related to the phenomenon of water penetration into the cellular structure of the samples due to disequilibrium processing (as in the extraction process where ultrasound is widely used). A higher water content of the samples after a 60 min treatment compared to a 30 or 45 min treatment may significantly increase the drying time, and there is no significant difference between samples treated for 45-min and 60 min, regardless of amplitude. Figure 2 shows the comparison of drying time for untreated samples and samples treated with the optimum processing parameters. It is evident that both constant rate and falling rate periods were affected by the ultrasonic treatment, confirming previous claims about the effects of cavitation on the matrix, changing the microstructure, and increasing mass transfer rates. The optimal parameters determined for ultrasonic treatment are consistent with the results of other studies on ultrasonic drying. Soquetta et al. show that ultrasonic pretreatment significantly changes the drying time of beets due to the influence of the released mechanical and thermal energy on the structure of the beets [46]. Jarahizadeh et al. showed that the application of ultrasound has a significant effect on the constant rate period during drying, which is also related to the increase in the mass diffusion rate due to the enlarged pores caused by cavitation [49]. This phenomenon, caused by cavitation bubbles, results from the formation of microchannels and potential changes in the cell membrane and proves to be interesting when large amounts of water need to be removed from pumpkins or other fruits and vegetables [50].

3.3. Artificial Neural Networks

After testing different transfer functions, both neural networks were obtained using multilayer perceptron with the BFGS training algorithm. The ANN-1 network for optimal parameters (90% for 45 min) consists of 10 hidden neurons, with a validation performance of 0.999991. The mean square error was 9.02×10^{-6}. It is evident that the MSE was much lower compared to the Weibull model, as shown in Figure 4, indicating a better fit than any of the analyzed mathematical models. This is consistent with studies by various authors who have compared ANN with empirical models [51,52].

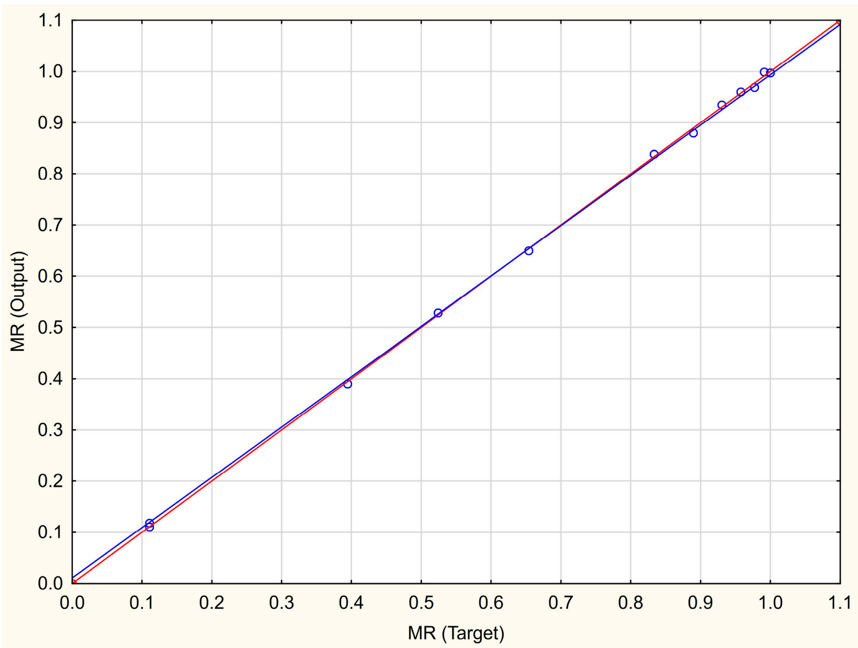

Figure 4. Comparison of fitting of ANN-1 and Weibull model to observed vs predicted values.

The ANN-2 network, consisting of 15 neurons in hidden layers, had a performance of 0.999987. The mean square error was 8.03×10^{-5}. It can be concluded that artificial neural networks can be successfully used to model the drying process in combination with ultrasound pretreatment. Moreover, both ANNs (Figures 4 and 5) were significantly more accurate than the standard empirical models tested. It should be taken into consideration that empirical models such as Weibull were based on only one set of process parameters, while the significantly better ANN-2 network considered all process parameters as inputs. This shows that ANN-2 can successfully predict the effects of all tested process variables on the drying time/moisture ratio, thus eliminating the need for multiple models that could become useless even with minor changes in the drying process. Due to the minimal errors and $R^2 > 0.999$, both ANN models can replace mathematical models for ultrasonic drying of pumpkin slices, regardless of the process parameters used.

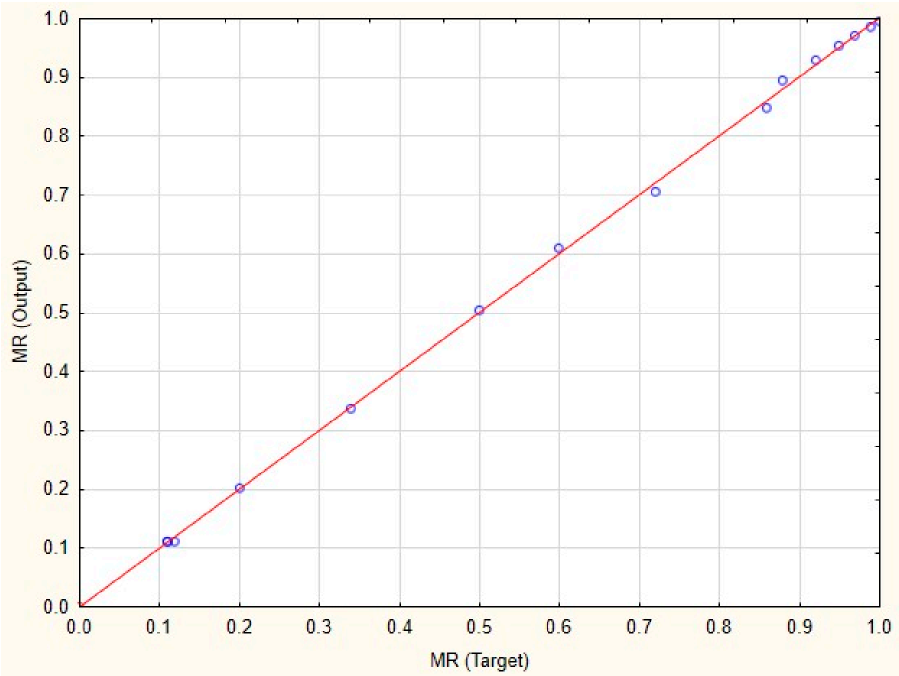

Figure 5. Fitting of ANN-2 to experimental data.

4. Conclusions

The drying of pumpkin pulp using ultrasonic pretreatment shows that increasing processing power by up to 90% significantly reduces drying time. An increase in the ultrasonic processing time negatively influences the drying process, so that prolonging drying time beyond 45 min only leads to increasing energy costs. The performed analysis of the mathematical models shows that the Page model has the best fit for drying the untreated samples, while the Weibull model has the best fit for the ultrasonically pretreated dried samples. This model can further be used as a basis for estimating drying parameters and potentially for the design of ultrasonic drying processes or equipment. Color change of samples was evident, but minimal. The lowest color change was obtained using optimal processing parameters. However, the best fit to the process parameters was obtained using the artificial neural network, which was found to be more accurate in predicting the effects of process parameters on the drying process.

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