Assessing the Quality of Lye-Peeled Garlic: A Concentration-Dependent Approach with Artificial Neural Networks and Multivariate Analysis

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Abstract— *The effectiveness of lye-peeled garlic was evaluated based on its chemical characteristics: allicin content, pyruvic acid, total phenolic content (TPC), total flavonoid content (TFC), antioxidant activity (AA), and anti-nutritional properties. The garlic cloves were peeled using the lye solutions (2–16%) in a ratio of 1:5 at 40±2 °C exposed for 7 min. The principal component analysis (PCA) was used to develop to assess the effectiveness of lye-peeling on the quality parameters. In addition, a two-layered feed-forward artificial neural network (ANN) was applied to develop a model to assess the quality parameters of peeled garlic. The found regression correlations 1.00000, 0.99572, and 0.99054 found for training, validation, and testing of artificial neural networks reflected the applicability of the developed model in quality assessment as regression performance was found to be 0.99005. It is concluded that 12% lye solution for peeling garlic cloves provides better quality, having less unpeeled garlic surface area. Therefore, this research highlights the application of lye concentration along with suitable temperature for garlic peeling with affecting the phytochemicals and nutritional quality of garlic*.

Keywords— Allium sativum, Artificial neural network, Lye peeling, Allicin, Antioxidant activity, Anti-nutritional property.

I. INTRODUCTION

Garlic (*Allium sativum* L.) is a bulb crop containing 2-4 layers of concentric semi-thick curvy masses known as cloves. The centre of garlic has a woody stem. It is grown worldwide, and 90% of garlic production is restricted to the Asian subcontinent, particularly in China and India (FAO, 2021). Garlic is mainly used for culinary purposes as a thickening agent in food preparation. It is also used to produce various processed products, namely pickles, paste, flakes, grits, powder, and so on. Garlic has a specific aroma, so it is also used to prepare mayonnaise, sausages, ketchup, and stews (Prasad *et al*., 2002). Its demand has increased several-fold because of its potential roles, i.e., as a flavouring agent, thickening agent, antimicrobial, therapeutic and medicinal properties. Recently, various researchers have proven that it reduces arterial pressure, controls platelet aggregation, inhibits cancer cells, and controls the cholesterol level in the blood (Pardo *et al*., 2007; Rekowska and Skupień. 2009; Shekhar *et al*., 2023).Since antiquity, raw garlic paste was traditionally combined with turmeric and/or onion to help relieve pain and sprains. These health benefits may be due to the occurrence of various types of volatile and non-volatile compounds. S-alk(en)yl-L-cysteine sulfoxide (alliin), which is present in the cytoplasmic vesicles at approximately 1%, is one of the significant compounds associated with raw garlic (Amagase *et al*., 2001). When raw garlic is crushed, the alliin reacts with the alliinase enzyme and forms allicin, an active unstable compound containing 60-80% of total thiosulfinates (Block, 1992). Other compounds, such as ammonia and pyruvic acid, are also formed during hydrolysis. Pyruvic acid, a stable compound, is correlated with the pungency of garlic. Although garlic contains a high concentration of bioactive compounds, it also contains a small quantity of anti-nutritional compounds, particularly tannins, saponins, phytic acid, flavonoids, and steroids (Yusuf *et al*., 2018).

Garlic is generally peeled before its use and mainly hand-peeled with the help of a knife. This method is laborious, time-consuming, expensive, and questionable regarding the uniformity of quality parameters. At the same time, with the change in lifestyles, there is an increase in the demand for either minimally processed products or semi-processed products for time-saving and ease of use for intended purposes. Due to this, peeled garlic demand on an industrial and home scale increased massively.

Lye peeling is one of the unit operations used for peeling of fruits and vegetables (Ramaswamy and Marcotte 2006). Garlic peel is rich in pectin, lignin, cellulose, and hemicellulose. Pectin acts as a binding agent, and lignin, an aromatic biopolymer, tightly binds cellulosic and hemicellulosic compounds. NaOH (10%)solution reduces the pectin and lignin content by breaking down or dissolving these compounds (Kumar *et al.*, 2022). It also dissolves the inner epicuticular wax by penetrating the epidermis layer, digests the cell walls and middle lamella, and loosens the thin membrane skin (Gould, 2013). Lye peeling may assist in removing potato skin and can hinder the enzymatic browning reaction by decreasing the reducing sugar concentration (Hidayat and Setyadjit 2019).

Dynamic model behaviour in the processing of some specific agricultural commodities using artificial neural networks (ANN) has gained momentum due to the ability to learn the neural networks. It is suitable for identifying complex agricultural product quality responses that mathematical approaches do not solve quickly. The ANN model was tried to dry sliced potatoes and fresh green peas to predict moisture content and residual vitamin C (Kamiński *et al.*, 1998). At the same time, the PCA technique was used for the optimization of microwave roasting of black rice based on a developed correlation between the resultant parameters, namely physical-functional properties, thermal properties, antioxidant and anthocyanin content (Arora *et al.*, 2021).

The present research was planned to study the effects of lye concentrations on garlic peeling without affecting the quality attributes, namely chemical parameters, pungency factors, antioxidants, and anti-nutritional properties. A quality parameters prediction model was developed to assess the effectiveness of lye peeling behaviour using ANN. PCA was applied to position the lye-peeled and manually peeled garlic based on a developed correlation between the measured parameters and applied lye concentrations.

II. MATERIALS AND METHODS

2.1 Materials:

Freshly harvested garlic bulbs (Haryana garlic-17) were brought from CCS-HAU, Hisar, Haryana. Manually, garlic cloves were separated from the whole bulbs. Mature garlic cloves of similar size were used for all the experimental work. L-cysteine,5,5'-dithio-bis-(2-nitrobenzoic acid) (DTNB),(4-(2-hydroxyethyl)-1-piperazineethanesulfonic acid (HEPS), 2,4 dinitrophenylhydrazine (DNPH), and sodium pyruvate were purchased from Nice Chemicals Pvt. Ltd., (India). Other chemicals utilized in the current research work were of analytical grade.

2.2 Lye peeling of garlic:

The garlic cloves were divided into 27 groups to conduct independent experiments in triplicate. Each group contained approximately 30 g of samples for manual or lye peeling. The garlic cloves were immersed in lye solution having concentrations of 2% (LP2), 4% (LP4), 6% (LP4), 8% (LP8), 10% (LP10), 12% (LP12), 14% (LP14), and 16% (LP16) for the lye peeling considering manual peeling as control. The tangible unpeeled garlic surface area was estimated with the help of transparent paper and converted into percentages (Oladejo *et al.*, 2014). The lye solutions were freshly prepared for each treatment. The temperature was maintained 40±2°C throughout the lye peeling process, i.e., 7 min. Immediately after the lye solution treatment, the left garlic peel was removed by rubbing the cloves under tap water. The surface moisture was removed using tissue paper. The phenolphthalein indicator test was performed for the presence of lye solution. The peeled garlic cloves were further used for different analyses.

2.3 Juice extraction process:

Around 17 g of peeled garlic clove was taken for juice extraction. It was crushed using the pestle and mortar without adding water. The juice was collected by pressing the garlic paste using a nylon cloth. The obtained juice was immediately used for the estimation of chemical properties.

2.4 Chemical properties analysis:

The pH of garlic juice was estimated using a previously calibrated electrical pH meter (model: LMPH-10, make: Labman Scientific Instruments Pvt. Ltd., India) at 20°C. Total acidity was estimated by the titration method (AOAC, 2006). Total soluble solids (TSS) of the garlic juice sample (20°C) were measured directly with a digital refractometer (model: 3810 PAL-1 digital, make: Atago Co. Ltd., Japan). The amount of reducing sugar in the garlic sample was estimated (Ahmad, 1996). The absorbance was taken at a 440 nm wavelength with the help of a UV-Vis spectrophotometer (model: DR6000, make: HACH company, USA). It was calculated using a standard glucose solution as a reference curve and expressed in g/kg. Ascorbic acid (AS.A) in the garlic samples was estimated using the 2,6-dichlorophenol indophenol reagent titration method with slight modification (Manas, 2014). Approximately 1 g of garlic was taken to prepare the extract using 25 ml of 4% oxalic acid. It was centrifuged at 8000 rpm for 18 min. Then, 10 ml of aliquot and 10 ml of 4% oxalic acid were titrated against 2,6-dichlorophenol indophenol reagent up to the appearance of a faint pink colour. Further, 5 ml of 0.1 mg/ml standard L-ascorbic acid solution and 10 ml of 4% oxalic acid were taken and titrated against 2,6-dichlorophenol indophenol reagent. The ascorbic acid (mg/g) concentration was calculated by the following equation.

$$Ascorbic\ acid\ = \frac{\text{Concentration}_{\text{standard solution}} \times \text{Titre volume}_{sample} \times \text{Volume taken for titration}}{Titre\ volume_{\text{standard solution}} \times Total\ volume\ made \times Wt\ of\ sample} \quad (1)$$

2.5 Allicin and Pyruvic acid content analysis:

The allicin and pyruvic content of the manual and lye-peeled garlic samples were measured by the spectrophotometric method at the wavelength of 412nm, and 420 nm, respectively (Prakash and Prasad, 2023a).

2.6 Total phenolic content (TPC), Total flavonoid content (TFC), Antioxidant activity (AA) analysis:

A solvent extraction technique was used to extract the garlic extracts (Prakash and Prasad, 2023a). TPC in the garlic extract samples was measured using a UV-spectrophotometer at 765 nm wavelength with minor modification (Prakash and Prasad, 2023a). The colourimetric technique was used to estimate the TFC (510 nm) in the garlic extract sample with slight modification (Bhandari and Rajbhandari, 2015). The DPPH antioxidant activity of the garlic sample was estimated by the method reported (Kinalski and Noreña, 2014).

2.7 Anti-nutritional properties:

Tannin, saponin, and phytic acid content in the garlic samples were estimated by the method followed by Prakash and Prasad (2023b).

2.8 Artificial neural network (ANN):

ANN is one of the malleable computational architects drawn by the inspiration from the human brain. It is capable enough for the modelling of any system or process before they are trained with the supply of sufficient numeric data obtained from the experiments. ANN tools can be implemented in a complicated model with numerous inputs and outputs data with a little experimental work. This type of model may be applied in the prediction of experimental data. The conditions chosen for garlic peeling were well-suitable for ANN model because of its non-linear attributes. In the present study, Matlab R2021 was applied for the ANN modelling. Manual peeling (MP) and different concentrations of lye peeling of garlic cloves were modelled with two-layer feed-forward neural networks. Neural network mapping was developed between numeric input data (peeling conditions) and numeric target data (quality parameters). The manual peeling of numerical input data was noted by the notation 0 (zero). The lye concentration of 16% solution was kept separate for the performance test of the developed model. The input and target data were statically represented in the [1×8] and [13×8] matrices, respectively. In the matrix, the first number represents the parameters, and the second represents the samples. The samples were randomly divided into 3 data sets, which were further used in training (50%), validation (25%), and testing (25%) functions. The training data set was used in the neural network training, and accordingly, its network error was adjusted. A validation data set was employed to assess the network's generalization, and when generalization reached a certain point, stop the training. Training did not affect the testing data set and offered a reliable network performance indicator before and after training.

The neural network contained one input layer, one hidden layer having ten neurons and thirteen output layers (Figure 1). Here, ten neurons were selected for the best prediction of results (outcomes) since a lower integer of neurons may not adequately cover the entire required network in the training step, while larger integers might memorize the network training patterns. Multiple hidden layers were used for a complex and particular application, but one hidden layer was sufficient for continuous non-linear functions (Mitra *et al*., 2011). The network was trained with the Levenberg-Marquardtback-propagation algorithm

(Fabani *et al.*, 2021). The training ceased automatically when generalization stopped improving the validated data set's mean square error (MSE). The training was performed until the mean square error (MSE) value was minimized to near zero, and the regression correlation (R) value maximized near one. Then, the performance check executed a sensitivity analysis against the developed model. The final ANN outcomes data were correlated with experimental data.

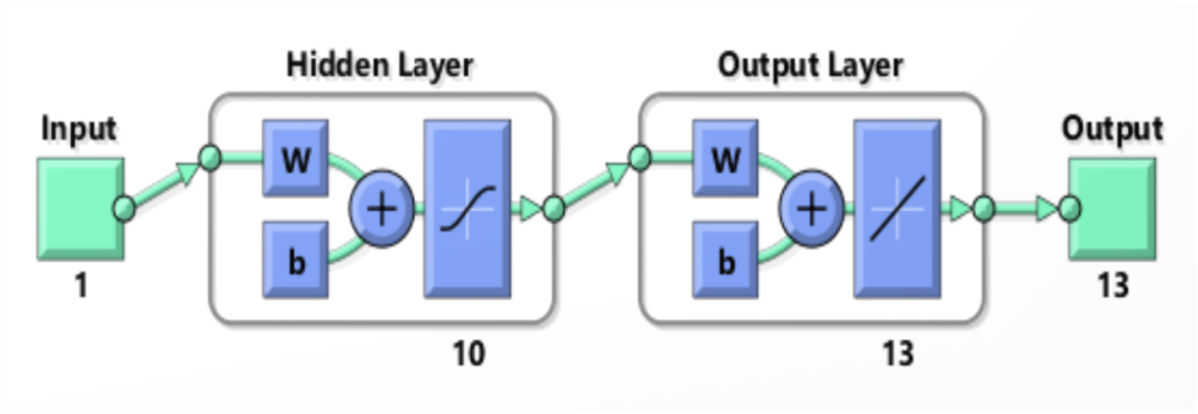

FIGURE 1: An ANN diagram of garlic lye peeling in different concentrations

2.9 Statistical analysis:

All the experimental parameters were performed in triplicate. The mean value was calculated using Microsoft Excel version 2007. Principal Component Analysis (PCA) was executed on mean data to explain the correlation between the variability of estimated parameters of the manual and different concentrations of lye-peeled garlic.

III. RESULTS AND DISCUSSIONS

3.1 Effect of lye concentration on garlic peeling:

The tangible unpeeled garlic surface area was reduced as the concentration of lye solution increased (Figure 2). At LP12, the garlic peeling rate was found to be maximum. It might be because the penetration power and diffusion process reach maximum at the exposed concentration and temperature (40°C). It reduces the amount of pectin (76.00%) and lignin (25.91%) content by diffusion of these compounds in 12% lye solution. The remaining peel content was mainly cellulose and hemicelluloses, which were easily removed by the cloves with gentle rubbing in water. The lye peeling efficiency process was influenced by the concentration of the lye solution, temperature, and immersion time (Caceres *et al.*, 2012).

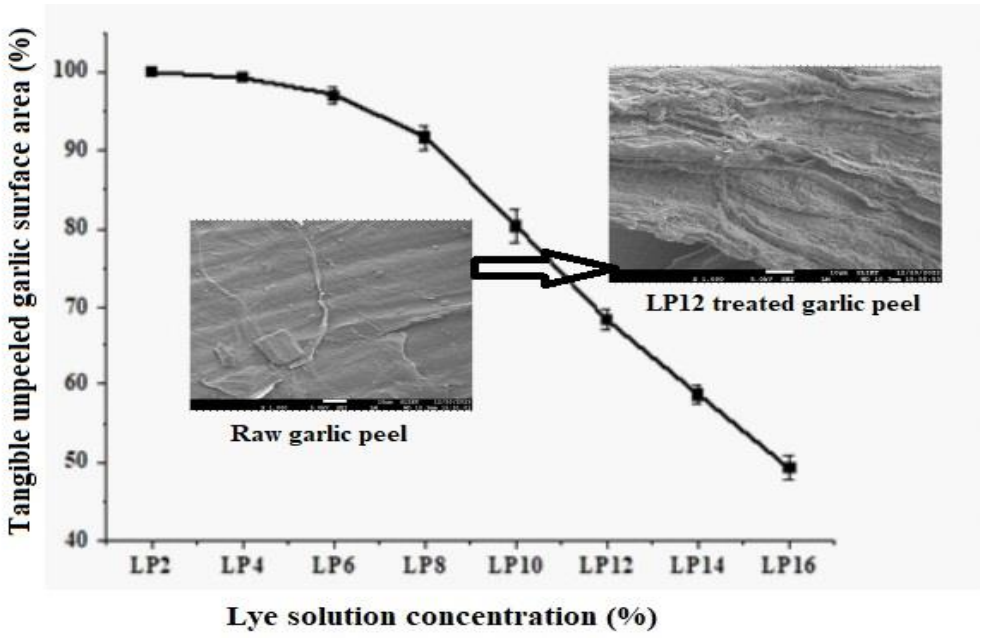

FIGURE 2: Effect of lye concentrations on tangible unpeeled garlic surface area

3.2 ANN model and its performance evaluation:

Training and validation phases are essential for developing the best ANN model. In the training phase, repeated iteration was used to achieve a regression coefficient near or equal to one. The best ANN model was found after 17th iteration, where regression coefficients for training, validation, and testing were 1.00000, 0.99572, and 0.99054, respectively (Figure 3A). Most errors were found in the testing stage because these datasets were not utilized in the training for the development of ANN network.

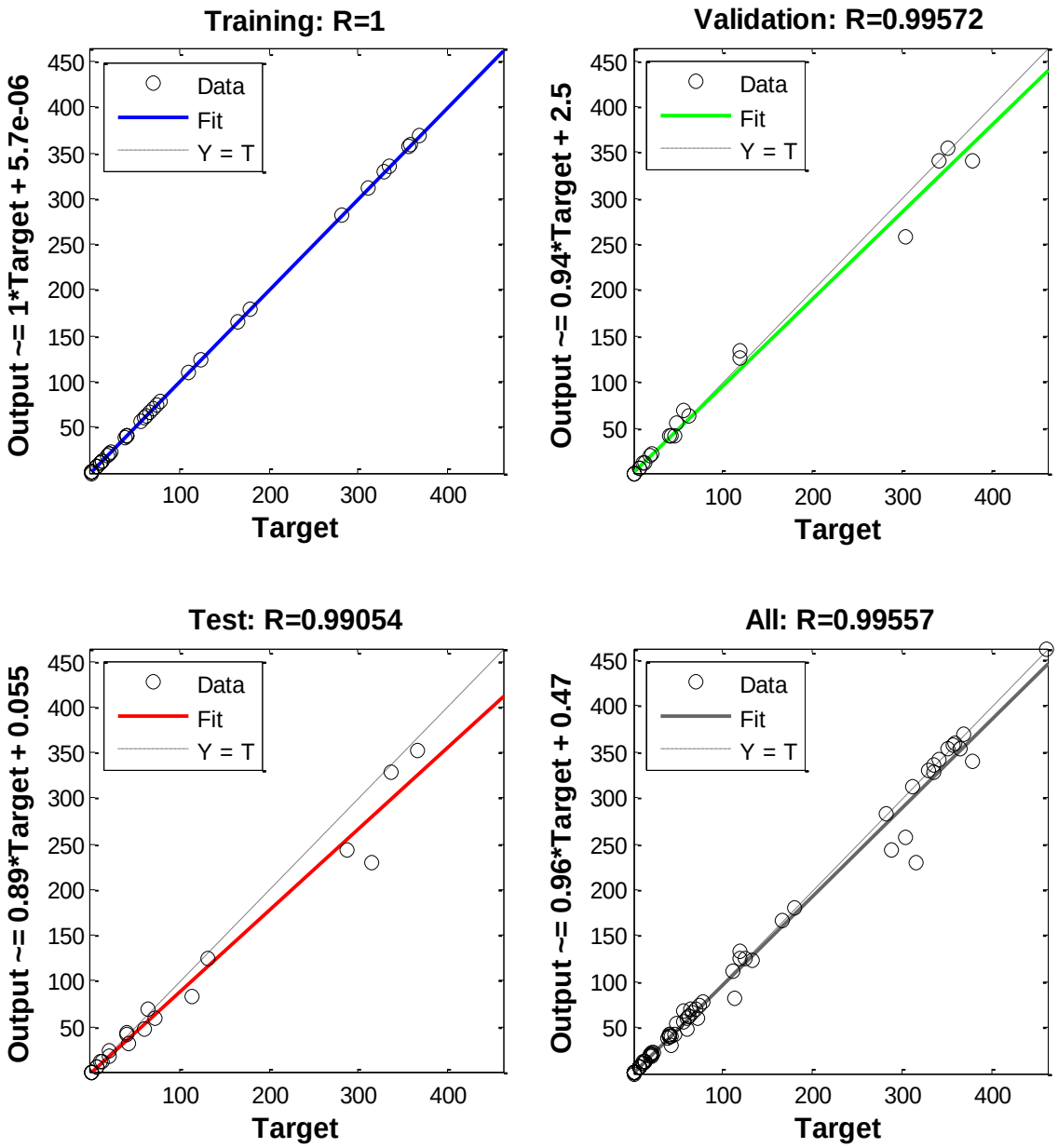

FIGURE 3A: Regressions plot in ANN trained model (Matlab R2013a software)

Once the ANN model was developed, its performance test was conducted with separate experimental data for practical application. The performance test data showed a regression coefficient of 0.99005, like the test data regression coefficient (Figure 3B). The developed model may thus be applicable in effectively predicting garlic peeling at different concentrations of lye peeling.

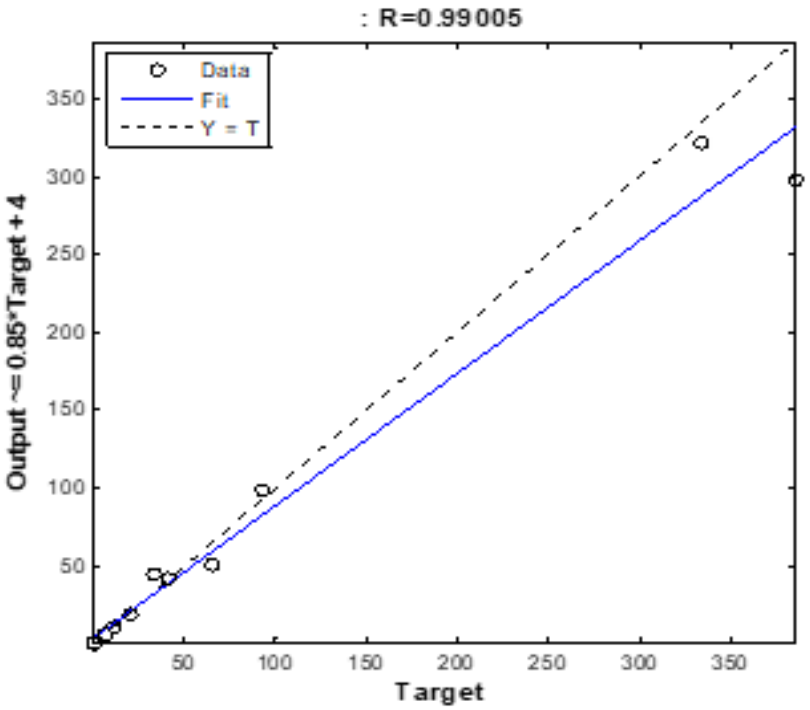

FIGURE 3B: Regression performance test of the developed neural network model (Matlab R2013a software)

3.3 Comparison of quality attributes of experimental and predicted ANN model outcomes:

The patterns of the experimental quality attributes and prediction by ANN model data set for chemical properties, bioactive compounds, i.e., allicin and pyruvic acid, TPC, TFC, AA, and anti-nutritional properties of lye-peeled garlic, are shown in Figures. 4 to 7, respectively. The remarkable point in the graphs is the pattern of the experimental and ANN model predicted results at different concentrations of peeled garlic. It indicates that ANN data outcomes are appropriately fitted and not under or over-fitted. Besides, it also noticed that the highest differences (errors) occurred at 4% and/or 10% of lye-peeled garlic in all the measured parameters. It might be because these concentrations of lye-peeled garlic were not involved in the training of the ANN model, and these data points were used to test the results. It means that the ANN model's outcome errors may be improved by training with the same experimental data sets or by minimizing the experimental errors at the same experimental points.

3.4 Chemical properties analysis:

The pH (6.10–6.23) and TSS (40.17–41.83°Brix) were varied as the concentration of lye solution increased from 2 to 14% for the lye peeling of garlic. The MP method had a pH similar to the lye-peeled garlic at 2, and 4% lye solutions, and other solutions had a slightly higher pH. Peeled garlic obtained from the MP method had less TSS content than all the lye-peeled garlic concentrations. Total acidity varied from 0.757 to 0.509% as the concentration increased from 2 to 14% (Figure 4). These changes might be due to the penetration power differences at different concentrations. At lower lye solutions, penetration power was almost negligible, and later, it was increased (Figure 2). Ascorbic acid content was decreased in all the concentrations of lye-peeled garlic (0.126–0.089mg/g) compared to MP garlic (0.142mg/g). Similar results were found in kiwi fruits, peeled with the concentrations of caustic solutions, i.e., 13, 18, and 23%, respectively (Guldas, 2003). Its ascorbic acid content decreased from 89 to 84.5% as there was an increase in caustic solution concentration. Manually peeled garlic had the highest amount of reducing sugar, and the lowest amount was found in a 2% lye solution. As the concentration of lye solution increased (2 to 14%), reducing sugar also increased but remained substantially less than the MP garlic (Figure 4). Pomelo peel was treated with various concentrations of alkaline (NaOH) solutions, and it was found that the reducing sugar increased from 52.81 to 91.01 g/100g (Chatkaew *et al*., 2021).

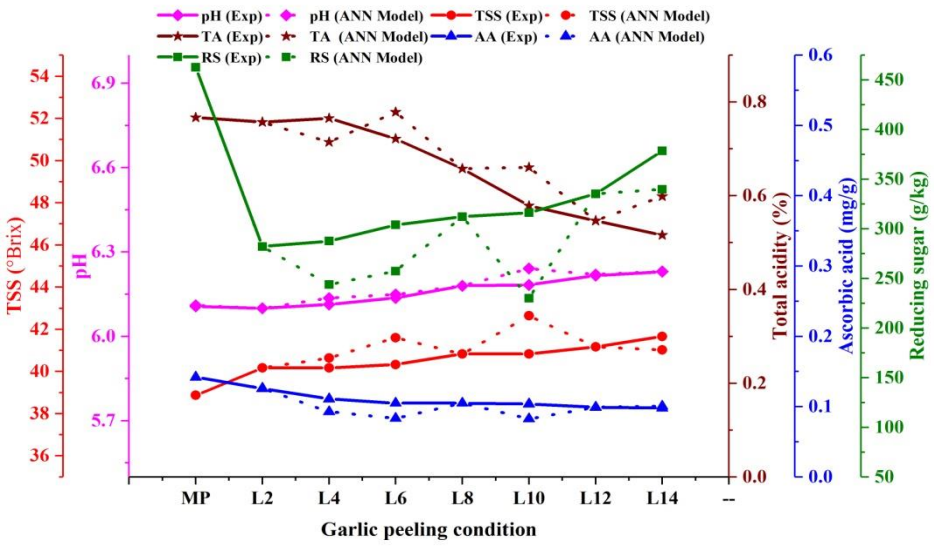

FIGURE 4: Experimental and ANN model predicated data of chemical parameters (manual peeled and lye-peeled garlic)

3.5 Allicin and pyruvic acid content analysis:

The allicin and pyruvic acid content in MP garlic and the different concentrations of lye-peeled garlic are shown in Figure 5. The manually peeled garlic contained 369.84 mg/100g of allicin and 69.40 µmol/ml of juice of pyruvic acid. Allicin content is relatively unstable at higher pH (Lee *et al*., 2014) or retard the chemical process; it may be the significant reduction of allicin and pyruvic acid content in lye solution peeled garlic. The lye-peeled garlic contained 330.13 to 366.19 mg/100g of allicin and59.77 to 66.51 µmol/ml of juice of pyruvic acid. The maximum amounts of allicin and pyruvic acid were present in peeled garlic with a 12% lye solution. Pyruvic acid content was lower (30.28µg/ml) in garlic pulp when garlic pulp was heated under ohmic conditions (Poojitha and Athmaselvi, 2020).

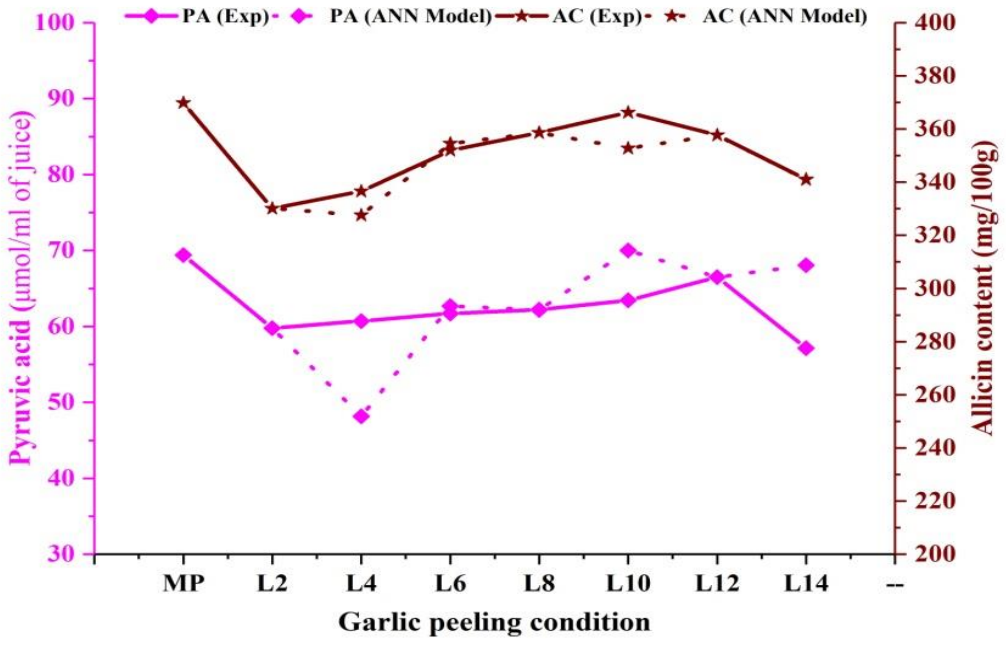

FIGURE 5: Experimental and ANN model predicated data of pyruvic acid and allicin content (manual peeled and lye-peeled garlic)

3.6 TPC, TFC, and AA properties analysis:

The TPC, TFC, and AA of different concentrations of lye-peeled garlic are presented in Figure 6. The TPC and TFC of lye-peeled garlic ranged from 136.56–248.01 mg/100g and 314.56–471.79 mg/100g at 2–14% concentration of lye-peeled garlic, respectively. The maximum amount of TPC (260.00 mg/100g) and TFC (511.08mg/100g) of garlic was found in manually peeled garlic, followed by 12% lye-peeled garlic. These significant changes may be due to the alteration in the structure of garlic and the destruction of the peel and outer waxy layer during lye peeling (Carranza-Concha *et al*., 2012). DDPH radical scavenging activities ranged between 19.20 and 21.15%, while manually peeled garlic was 22.53%. However, in DDPH radical scavenging activities, some changes occurred to the TPC and TFC. It might be due to less correlation with other compounds, particularly TPC, TFC, and ascorbic acid. Several researchers stated that there was no correlation between these compounds (Carranza-Concha *et al*., 2012; Kuskoski *et al*., 2005).

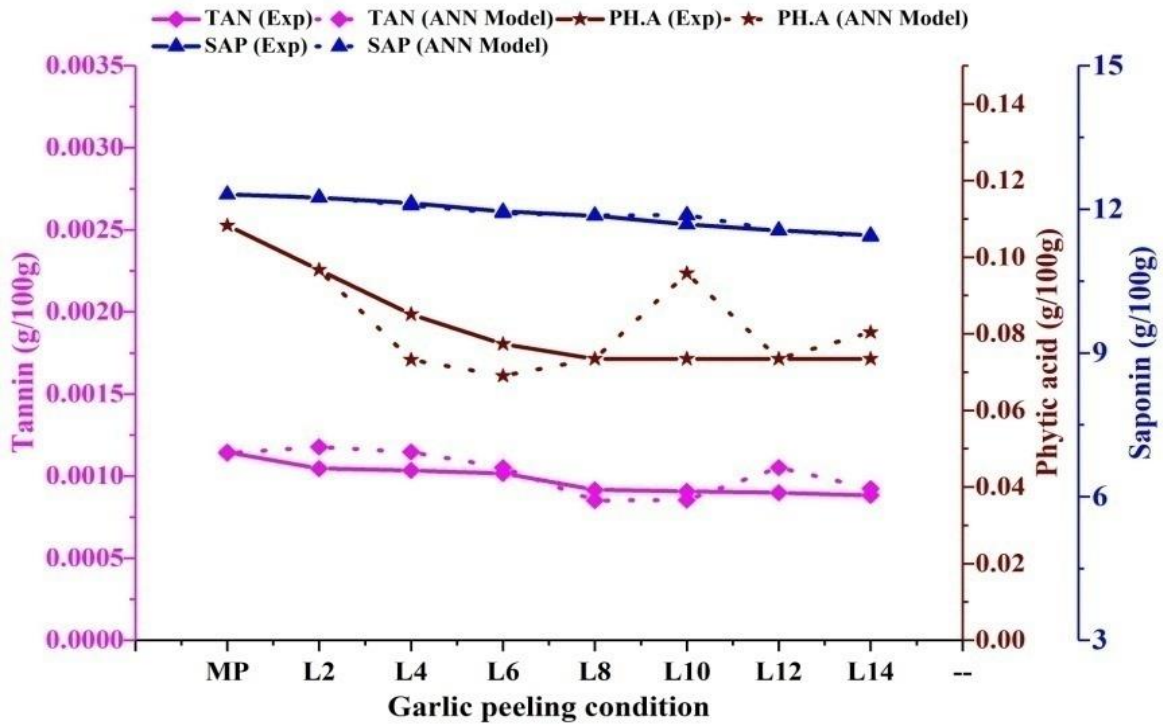

FIGURE 6: Experimental and ANN model predicated data of TPC, TFC and AA (manual peeled and lye-peeled garlic)

3.7 Anti-nutritional properties:

Tannins, astringent compounds belonging to polyphenols, combine with protein and other organic compounds to form a complex precipitate. Due to the formation of complex compounds, they block the nutritional content (Gemede and Ratta, 2014). Figure 7 shows that the tannins decreased by up to 15.65% when the concentration of lye solution increased from 2 to 14%. The tannins were reduced to 22.95% compared with the MP garlic over 14% lye-peeled garlic. Similar results were reported (Jyothi and Sumathi, 1995) on treating a low sodium hydroxide solution on common beans at room temperature for 24 hrs.

Phytic acid, an anti-nutritional compound, reduces the bioavailability of minerals to humans, primarily zinc, iron, calcium, magnesium, manganese, and copper (Gemede and Ratta, 2014). Phytic acid was found to be 0.1083g/100g in raw garlic (Figure 7). Its concentration decreased from 10.71 to 35.71% when the lye solution concentration increased from 2 to 14% for garlic peeling. The rate of phytic acid concentration drastically decreased in the initial stage of lye-peeled garlic; later, it decreased in the steady state. A similar phytic acid result was found in common beans when soaking in sodium hydroxide solution (14.6% in 24 hrs) at room temperature (Jyothi and Sumathi,1995).

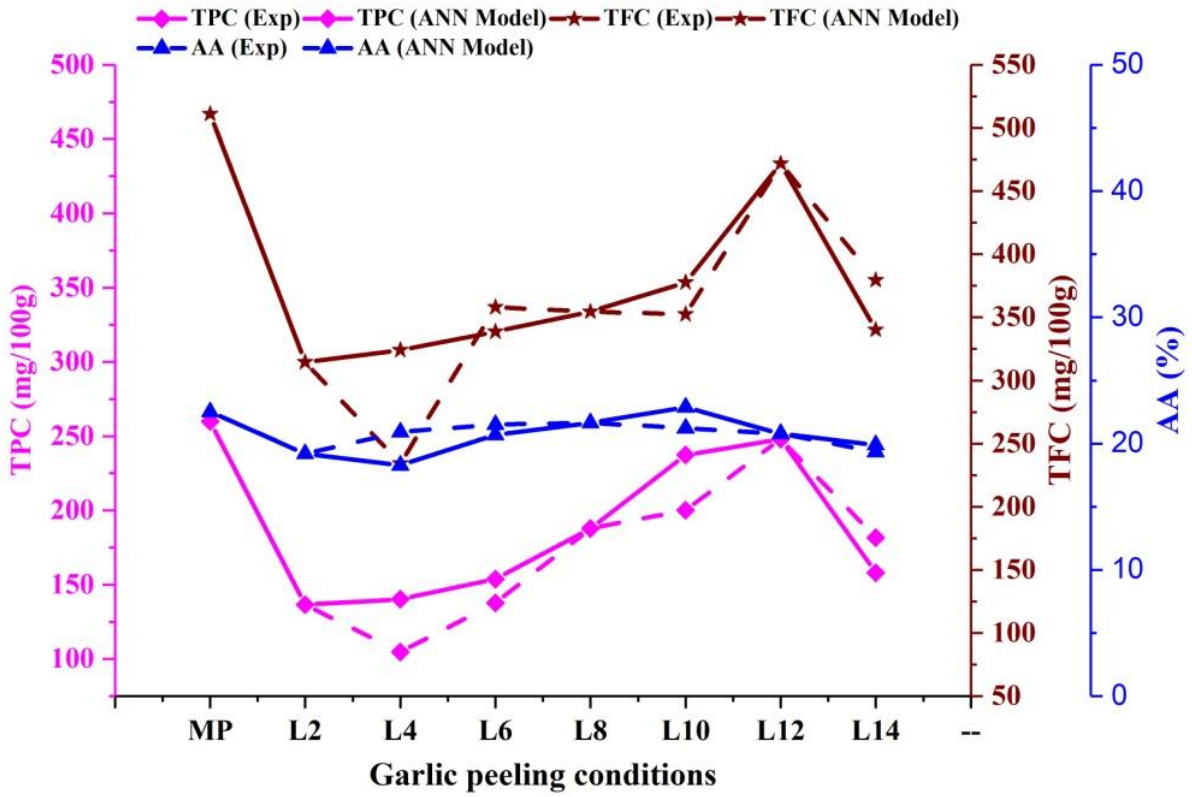

FIGURE 7: Experimental and ANN model predicated data of anti-nutritional properties (manual peeled and lye-peeled garlic)

Anti-nutritional compounds, namely saponins, can retard the absorption of some non-polar vitamins, minerals, and proteins due to the formation of insoluble complexes (Francis *et al.*, 2002). The saponin concentration was reduced due to garlic peeling, with a 2–14% concentration over MP garlic (Figure 7). The saponins that were present in the raw garlic were 12.31 g/100g. At 14% NaOH concentration, garlic saponin content decreased by up to 6.92% when peeled with NaOH solution. The saponins were gradually reduced, like that of tannin.

3.8 Principal component analysis (PCA) and optimization of lye-peeled garlic:

PCA was used to develop the correlation between the lye peeling concentration and chemical parameters (pH, TSS, titratable acidity, ascorbic acid, and reducing sugar), pungency parameters (allicin content and pyruvic acid), TPC, TFC, AA, and Anti-nutritional properties (tannins, phytic acid, and saponins). Figure 8 shows that the PC1 and PC2 showed61.55% and 26.81% variations, respectively. It was also observed that TPC, TFC, AA, AC, PA, and RS are positively correlated with MP, LP8, LP10, and LP12. It may be due to a minor effect on native measured parameters. However, LP2, PP4, and LP6 formed separate clusters having the properties of AS.A, PH.A, TAN, SAP, and TA parameters. It may be because lye peeling concentration did not affect these parameters, particularly anti-nutritional properties.

The lye peeling concentration of garlic was optimized using the ANN model and PCA with minimum unpeeled garlic surface area. The rate of unpeeled garlic surface area was found to be lower at a 12% lye concentration. From the ANN model, it was found that the estimated parameters of lye-peeled garlic had a lower error at 12% lye concentration. PCA also showed less effect on quality parameters and more on anti-nutritional properties. Overall, it was observed that LP12 has lesser effects on measured quality parameters, particularly TPC, TFC, AA, AC, PA, and RS, and a more significant impact on anti-nutritional properties.

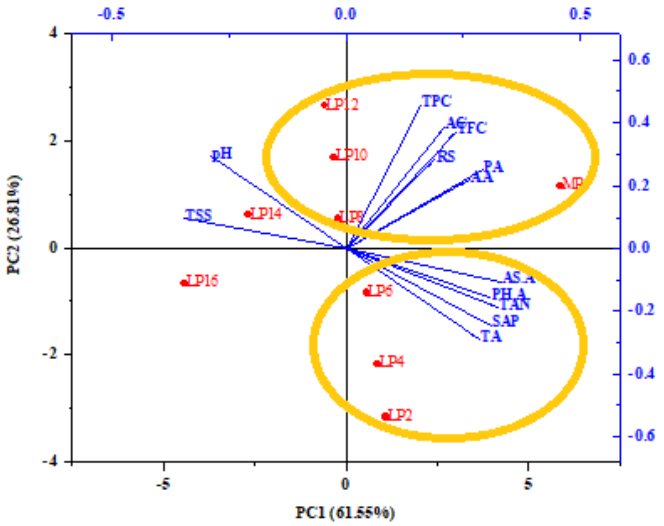

FIGURE 8: Biplot of PCA at different concentrations of lye peeling and manual peeling. Abbreviations: TS: total soluble solids; TA: titratable acidity; AS: ascorbic acid; RS: reducing sugar; AC: allicin content; PA: pyruvic acid; TPC: total phenolic content; TFC: total flavonoid content; AA: antioxidant activity (% reduction in 2,2-diphenyl-1-picrylhydrazyl hydrate); TAN: tannin; SAP: saponin; PH.A: phytic acid.

IV. CONCLUSION

Concentration-dependent lye peeling of garlic cloves was carried out, maintaining temperature and exposure time as a constant. The effective peeling was found with the 12% lye solution. At the optimized condition of peeling reducing sugar significantly reduced and helped in obtaining dehydrated garlic brighter without affecting other quality attributes. At the same time, anti-nutritional compounds, namely tannin, phytic acid, and saponin, decreased significantly. PCA showed that LP12 correlates better with TPC, TFC, AA, AC, PA, and RS, having a minimum unpeeled garlic surface area. The found regression correlations 1.00000, 0.99572, and 0.99054 for training, validation, and testing of ANN model reflected the applicability of the developed two-layered feed-forward ANN model in the process prediction in efficient manner. The application of ANN may be used to predict online garlic peeling and successfully estimate quality parameters at a commercial scale.

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CONFLICT OF INTEREST

The authors have no conflict of interest to declare.

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