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Fabrication of Al/ZnO electrode for e-tongue application using extreme learning machine

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ABSTRACT

Human body experiences sensation in five ways. They are the knowledge of sound, touch, sight, taste and smell. The knowledge of taste can be electronically experienced as e-tongue. The e-tongue imitates human test knowledge. To get the knowledge on the taste, there have been so many methods adopted by food processing, pharmaceutical and chemical processing industries. On further, they are not only expensive but also have incompetency of giving accurate taste like sweetness, sourness and saltiness through a single sensory bud. This work is intended for an analytical and experimental method to detect taste accurately. A set of the experiment has been carried out by using most promising bio-friendly material zinc oxide (ZnO). The taste concentration can be detected by using an electrochemical method. The chemical route method has chosen for the synthesis of ZnO solution. The modified chemical wet and dry technique (MCWD) is adopted due to uniform coating, inbuilt heat treatment, low cost and overall quick time to make a thin film on aluminium (Al) wire. To set an electrochemical setup, ZnO coated Al and silver (Ag) has taken as working and reference electrode respectively. To respond to different tastes like sour (citric acid), salty (sodium chloride) and sweet (glucose) the electrical data outputs from electrochemical reactions are analysed by varying respective concentrations in the aqueous media. For a sensory bud data validation, extreme learning machine (ELM) based single layered feed-forward neural networks (SLFNs) has been implemented to check the accuracy and pattern recognition.

Keywords:*E-tongue; Modified chemical wet and dry technique; Extreme learning machine; Single layered feed-forward neural networks.*

1. INTRODUCTION

The most important quality measures of the food and pharmaceuticals products are taste. However, we know the exact composition of any samplebutcannot tell anything about the taste of the sample, soanalytical measurements are not proper to address the problem.The electronic tongue isintended to measure a 'fingerprint' of the samples allowing sensitive comparison with the natural data which makes the technique suitable for taste measurements [1-3]. The taste of eatable products is solely responsible due to their chemical compositions and thus can be easily detected by the human tongue. For the Detection of dissolved organic and inorganic samples, there are seven sensory systems present electronically. Each sensor gives a reaction different from other. A combination of all sensors result generates a unique fingerprint pattern and thus makes the unique technique for taste measurement.

For the oral administration, the taste is an important parameter to decide the products intake. Most of the available medicinesare not good in taste. For the patient acceptability and compliance, the drugs are masked by adding different flavoring agents [4]. So,the taste assessment is one of the important quality control parameter for evaluating taste-masked formulations.An e-tongue comprises of the cell which is an electrochemical type, and can generate electrical energy from chemical reaction and pattern recognition system [5-7].This can be recognizedsoluble non-volatile molecules which form the taste of a sample. The e-tongue provides information about the evolution of bitterness intensity in function of time [8-10]. This can select the optimal formulation among the sensory probes having different coating thicknesses. E-tongue is used to taste sour, salty and sweet nature of the liquid sample [11, 12].

This study investigates the response of different tastes like sour (citric acid), salty (sodium chloride) and sweet (glucose) the electrical data outputs from electrochemical reactions are analyzed by varying respective concentrations in the aqueous media. For a sensory bud data validation, extreme learning machine (ELM) [13] based single layered feedforward neural networks (SLFNs) [14, 15] has been implemented to check the accuracy and pattern recognition. The remainder of this paper is organized as follows:

The proposed e-tongue experimental setup and ELM based pattern recognition model for the sensory bud data are described in section 2 and 3. The result, discussion and conclusion of this manuscript are discussed in section 4 and 5.

2. MATERIALS AND METHODS

2.1. E-tongue experimental setup.

The e-tongue experimental setup comprises electrodes/ probes (working and reference), hotplate, NI PXI 1042Q data acquisition system, pH meter with probe and sample container (50 ml beaker). The schematic diagram of the proposed e-tongue experimental setup is depicted in Figure 1. This is two probe cells used to study the electrochemical properties of an analyte in the solution.

In this work, Al and Ag wires have taken as a working and reference electrode respectively. For a sensing purpose, the working electrode is placed near to a reference electrode as close as possible inside the analytic solution. The potential of working electrode is not constant due electrochemical process on the working electrode surface. This deteriorates the sensing performance over the due course of time. The reference electrode is used to increase the sensing performance. A fixed and stable external potential of 0.75 V is applied for better sensing. At sensing electrode, the fixed voltage is maintained by the reference electrode throughout the process. The analytes are citric acid, glucose and sodium chloride.

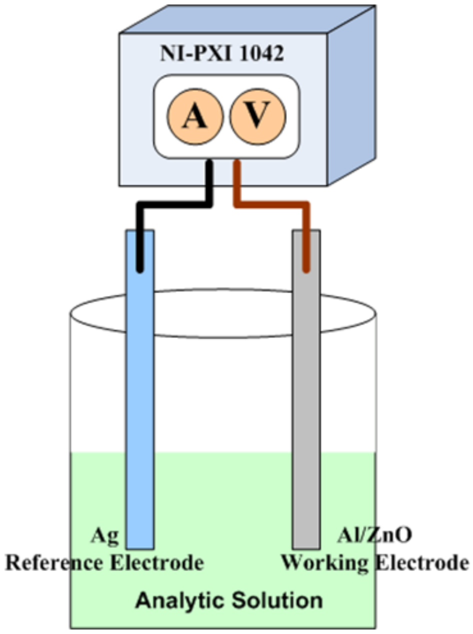

Figure 1. Schematic diagram of the proposed experimental setup.

Prior to experiment, the electrodes are cleaned. The cleaning procedure is a little bit difficult process to get a contamination free probe. In first attempt, the aluminum wire has cleaned by using acetone and dried in the oven at 50 ℃ for 10 minutes. In a petridis, 1ml of HCl and 2ml of distilled water are added and the dried probe is dipped inside the petridis. The petridis has kept in the ultrasonic bath for 20 minutes for ultrasonic cleaning. ZnO solution is prepared by adopting sol-gel technique. The MCWD has adapted to uniform coating especially for the inbuilt heat treatment, low cost and overall quick time for the making thin film on Al wire [12].

2.2. ELM based prediction analysis.

ELM theories extended to biological neurons whose mathematical formula is unknown [16]. Unlike conventional theories and common understanding, ELM can be made without tuning hidden neurons in wide type of neural network. The extreme learning machine was proposed in Huang et al [13]. The single layer feed forward network was trained with K hidden neurons and activation function g(x) to learn N distinct samples (x_i ,t_i), where xi = $[x_{i1}, x_{i2}, . . . , x_{in}]^T \in R^n$ and $t_i = [t_{i1}, t_{i2},\ldots, t_{im}]^T \in R^m$. The weights of the input layer and hidden biases are randomly generated in ELM. Applying this principle the nonlinear system has been converted to a linear system and thus depicted in equation 3.1.

$$H\beta = T \qquad (3.1)$$

Where H, the output hidden layer matrix corresponds to $\{h_{ij}\}$(i = 1,…,N and j = 1,……,K). The output of j^{th} hidden neuron is x_i and can be termed as$h_{ij} = g(w_j \cdot x_i + b_j)$. The weight vector, $w_j = [w_{j1}, w_{j2}, \ldots, w_j]^T$ is connected with j^{th}hidden neuron and input neuron. Bias of the system $b_j$denotes the bias of j^{th} hidden neuron and $w_j \cdot x_i$denotes the inner product of w_j and x_i. The output weight matrix and their connecting weight vectors of j^{th} hidden neuron with output neurons are $\beta = [\beta_1, \beta_2,\ldots, \beta_k]^T$ and $\beta_j = [\beta_{j1}, \beta_{j2},\ldots, \beta_{jm}]^T$ (j = 1, …, k). Whereas the target or desired matrix output is $T = [t_1, t_2, \ldots, t_n]^T$. The minimum norm least square solution to the linear system is shown in equation 3.2.

$$\hat{\beta} = H^{\dagger} T \qquad (3.2)$$

The Moore Penrose generalized inverse of H matrix is denoted as $H^{\dagger}$[17]. The unique and smallest norm among the least square solutions is $\hat{\beta}$. As analyzed in paper, ELM using MP inverse method increases the learning speed thus the performance. The pseudo code for the standard ELM algorithm is presented below:

1. The input weights w_i and b_i are randomly assigned ;
2. The output hidden layer matrix H is calculated, where H =$\{h_{ij}\}$(i = 1,…,N and j = 1,……,K) and $h_{ij}=g(w_j \cdot x_i + b_j)$;

Calculate the output weight matrix as $\hat{\beta} = H^{\dagger} T$, where $H^{\dagger}$ is the MP generalized inverse of matrix H.

3. RESULTS

The XRD analysis confirms the formation of the polycrystalline ZnO on Al which is already discussed in our earlier published paper. It is observed that some percentage of Al diffuses into the ZnO film at the interface region. In this work, four set of experiments have been carried out to sense different tastes. The inputs to the ELM model are the change in concentration and the current data values obtained from the probe at the time of experiment.In every experiment, the concentration of the solution changes from 10 to 100 µM with 10 µM step size. Figure 2 depicts the concentration of solution vs. current. The current values increase with an increase in concentration for citric and glucose values but in case of sodium chloride the current value decreases. There are three combined cases being verified and they are as follows: (i) citric acid + glucose, (ii) citric acid + sodium chloride and (iii) glucose + sodium chloride. In case of (i) and (ii) combination, no current was changed even if the concentration changes from 10 µM to 100 µM with an equal proportion of analyte 1:1.

Thus may happen due to the dominant behaviour of citric acid in the two cases. The combination of tastes like (sodium chloride and glucose) has also been verified by this method. The result values have depicted in figure 3. In this case glucose and sodium chloride have mixed at a proportion of 1:1. When the concentration of the analytic solution is 10 µM, at that time the individual concentration of glucose and salt is 5 µM each respectively. Similarly, at 70 µM analytic solution, the concentration of glucose and salt is 35 µM each respectively.Though, the salt has dominant nature over sour and sweet but in our case sweetness has a dominant impact when an equal proportion of salt and glucose added in distilled water. The result of which, the nature of curve shows as like glucose with lesser amount of current values than individual glucose solution.

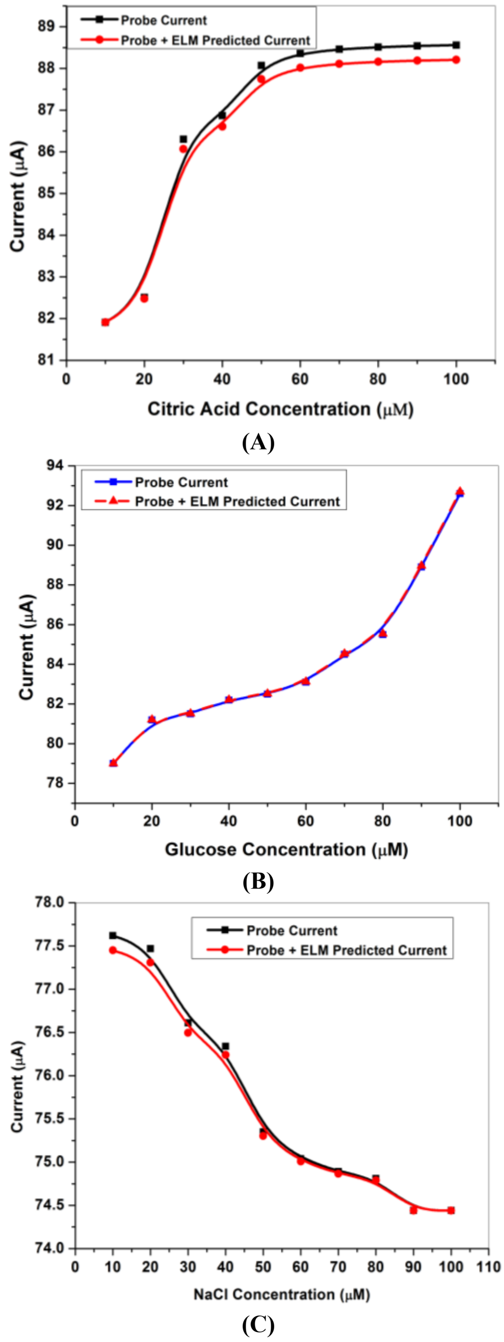

Figure 2. Concentration vs. current (A) Sour (Citric Acid), (B) Sweet (Glucose), (C) Salt (sodium chloride)

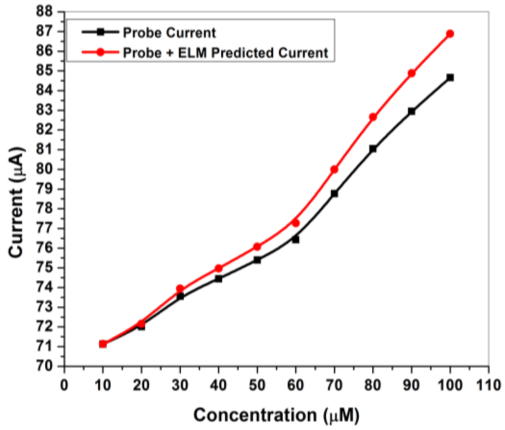

Figure 3. Concentration (glucose and sodium chloride at 1:1 proportion) vs. current.

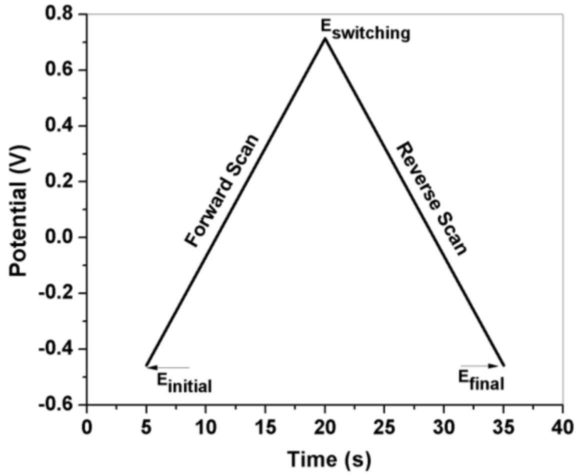

Figure 4. Excitation signal for cyclic voltammetry.

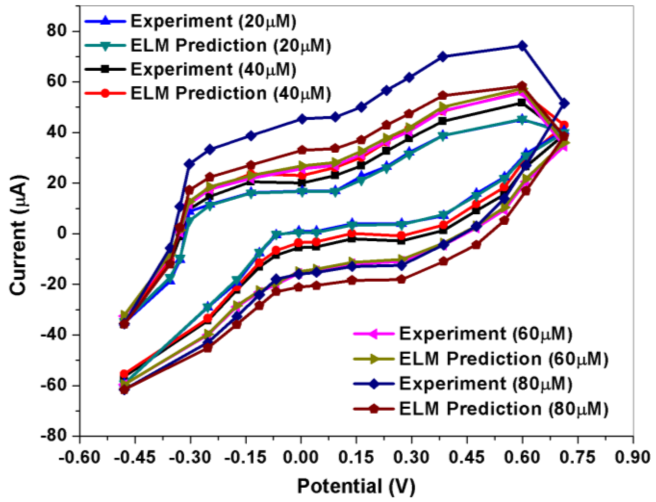

Figure 5. The voltammetry curve of the probe.

Table 1. Errors and correlation coefficient between probe and ELM predicted data values.

Sl. No	Data Set	MAE	RMSE	MAPE	CC
1.	Citric Acid	0.0391	0.0436	3.5566	0.9997
2.	Glucose	7.1469e-5	8.9951e-5	0.0065	0.9999
3.	Sodium chloride	6.2733e-5	8.5077e-5	0.0057	0.9996
4.	Sodium chloride + Glucose	3.175e-4	3.972e-4	0.0289	0.9997
5.	Voltammetry Curve: 20 μM	0.0056	0.0060	1.3653	0.9995
6.	Voltammetry Curve: 40 μM	0.0094	0.0100	2.2983	0.9989
7.	Voltammetry Curve: 60 μM	0.0238	0.0260	5.8432	0.9978
8.	Voltammetry Curve: 80 μM	0.0710	0.0797	17.4385	0.9966

The excitation signal for cyclic voltammetry is a linear potential scan with a triangular wave form and thus depicted in figure 4. The triangular excitation signal varies the potential of the electrode between two values, which cause the potential to scan positively from -0.45 V to 0.75 V and then causing a negative scan to come back to its original value. Figure 5 depicts the voltammetry curve/ the voltammogram of the probe at different concentration levels of the sodium chloride and glucose values vs. current.

This is a display of voltage vs. current curve. The current is measured at the working electrode during excitation signal and can be considered as a response signal to the potential excitation signal [18]. In many areas of chemistry application, the cyclic voltammetry has widely used for electro analytical technique for the study of redox process to understand the reactions and their intermediates [19-21]. In this experiment two probes are being used to monitor the current (I) as a function of applied potential (E) to generate the voltammetry curve. The combination of tastes particularly glucose and sodium chloride mixed with a proportion of 1:1 and at that mixed analyte being analysed under this electrochemical technique.

The table.1 depicts the errors like mean absolute error (MAE), root mean square error (RMSE), mean absolute percentage error (MAPE) and correlation coefficient between the probe produced data values and ELM response. The response like errors and correlation coefficient validates the probe data values and thus recognizes the signal pattern.

4. CONCLUSIONS

In this manuscript, we propose an e-tongue based on Al/ZnO to respond to different tastes like sour, salty and sweet. To create sourness, saltiness and sweetness in laboratory grade some chemical compounds are like citric acid, sodium chloride and glucose added in distilled water respectively. The MCWD has used to fabricate the probes and the electrochemical setup has proposed to sense the different aforesaid tastes. The sensing data values are stored in NI PXI 1042 labview data acquisition systems.

For a sensory bud data validation, extreme learning machine (ELM) based single layered feed-forward neural networks (SLFNs) has been implemented to check the accuracy and pattern recognition. The low cost, easy fabrication process and result accuracy data value enable the system uniqueness. The system with ELM based system can help to design industrial grade sensing system.

5. REFERENCES

1. Yusuke, T.; Toko, K. Electronic tongues–a review. *IEEE Sensors Journal* **2013**, *13*, 3001-3011, https://doi.org/10.1109/JSEN.2013.2263125.
2. Ouyang, Q.; Yang, Y.; Wu, J.; Chen, Q.; Guo, Z.; Li, H. Measurement of total free amino acids content in black tea using electronic tongue technology coupled with chemometrics. *LWT* **2020**, *118*, https://doi.org/10.1016/j.lwt.2019.108768.
3. Winquist, F.; Lundström, I.; Wide, P. The combination of an electronic tongue and an electronic nose. *Sensors and Actuators B: Chemical* **1999**, *58*,512-517, https://doi.org/10.1016/S0925-4005(99)00155-0.
4. Kulkarni, M.; Vishwakarma, B.; Sen, S.; Anupuram, S.; Date, A.A. Development and evaluation of taste masked dry syrup formulation of potassium chloride. *AAPS open* **2019**, *5*, 1, https://doi.org/10.1186/s41120-019-0030-z.
5. Krantz-Rülcker, C.; Stenberg, M.; Winquist, F.; Lundström, I. Electronic tongues for environmental monitoring based on sensor arrays and pattern recognition: a review. *Analytica chimica acta* **2001**, *426*, 217-226, https://doi.org/10.1016/S0003-2670(00)00873-4.
6. Banerjee, M.B.; Roy, R.B.; Tudu, B.; Bandyopadhyay, R.; Bhattacharyya, N. Black tea classification employing feature fusion of E-Nose and E-Tongue responses. *Journal of food engineering* **2019**, *244*, 55-63, https://doi.org/10.1016/j.jfoodeng.2018.09.022.
7. Palit, M.; Tudu, B.; Dutta, P.K.; Dutta, A.; Jana, A.; Roy, J.K.; Bhattacharyya, N.; Bandyopadhyay, R.; Chatterjee, A. Classification of black tea taste and correlation with tea taster's mark using voltammetric electronic tongue. *IEEE Transactions on Instrumentation and Measurement* **2009**, *59*, 2230-2239, https://doi.org/10.1109/TIM.2009.2032883.
8. Winquist, F. Voltammetric electronic tongues–basic principles and applications. *Microchimica Acta* **2008**, *163*, 3-10, https://doi.org/10.1007/s00604-007-0929-2.
9. Bleotu, C.; Mambet, C.; Matei, L.; Dragu, L.D. Improving Wine Quality and Safety Through Nanotechnology Applications. *Nanoengineering in the Beverage Industry* **2020**, 437-458, https://doi.org/10.1016/B978-0-12-816677-2.00015-6.
10. Vallejo, W.; Alvaro, C.; Díaz-Uribe, C. Methylene Blue Photodegradation under Visible Irradiation on Ag-Doped ZnO Thin Films. *International Journal of Photoenergy* **2020**, *2020*,11, https://doi.org/10.1155/2020/1627498.
11. Konchada, D.; Bhoi, B.B.; Pattanaik, P.; Mishra, B.;Kamilla, S.K. Preparation of TiO2 Film on Al by Sol–Gel Chemical Wet Dry Technique for Electrochemical Bio-Sensing Applications. *Advanced Science Letters* **2016**, *22,* 491-493, https://doi.org/10.1166/asl.2016.6895
12. Pattanaik, P.; Kamilla, S.K.; Das, D.P.; Mishra, D.K. Experimental and simulated study of electrical behaviour of ZnO film deposited on Al substrate for device applications. *Journal of Materials Science: Materials in Electronics* **2014**, *25*, 3062-3068, https://doi.org/10.1166/asl.2016.6895.
13. Huang, G.B.; Zhu, Q.Y.; Siew, C.K. Extreme learning machine: theory and applications. *Neurocomputing* **2006**, *70*,489-501, https://doi.org/10.1016/j.neucom.2005.12.126.
14. Huynh, H.T.; Won, Y. Application of Regularized Online Sequential Learning for Glucose Correction. *Transactions on Large-Scale Data-and Knowledge-Centered Systems XLI* **2019**, *11390*, 160-171, https://doi.org/10.1007/978-3-662-58808-6_7.
15. Cambria, E.; Gastaldo, P.; Bisio, F.; Zunino, R. An ELM-based model for affective analogical reasoning. *Neurocomputing* **2015**, *149,* 443-455, https://doi.org/10.1016/j.neucom.2014.01.064.
16. Zhu, Q.Y.; Qin, A.K.;Suganthan, P.N.; Huang, G.B. Evolutionary extreme learning machine. *Pattern recognition* **2005**, *38*, 1759-1763, https://doi.org/10.1016/j.patcog.2005.03.028.
17. Álvarez-Martos, I.; Henry, C.S.; Abedul, M.T.F. Anodic stripping voltammetric determination of lead and cadmium with stencil-printed transparency electrodes. In: *Laboratory Methods in Dynamic Electroanalysis.* Elsevier, 2020; pp. 35-45, https://doi.org/10.1016/B978-0-12-815932-3.00004-8.
18. Lima, T.S.; La-Scalea, M.A.; Raminelli, C.;Simões, F.R.; Franco, E.; da Silva, G.D.; Salvador, M.A.; Homem-de-Mello, P.; de Oliveira, H.P.M.; Codognoto, L. Voltammetric determination of chlorothalonil and its respective reduction mechanism studied by density functional theory. *Journal of Solid State Electrochemistry* **2019**, *23,* 553-563, https://doi.org/article/10.1007/s10008-018-4162-1.
19. Labbé, E.; Buriez, O. Fundamental Input of Analytical

Electrochemistry in the Determination of Intermediates and Reaction Mechanisms in Electrosynthetic Processes. *ChemElectroChem* **2019**, *6,* 4118-4125, https://doi.org/10.1002/celc.201900045.
20. Richter, J.B.; Eßbach, C.; Senkovska, I.; Kaskel, S.; Brunner, E. Quantitative in situ 13 C NMR studies of the electro-catalytic oxidation of ethanol. *Chemical Communications* **2019**, *55*, 6042-6045, https://doi.org/10.1039/C9CC02660F.
21. Miyajima, S. Enclosing Moore–Penrose inverses.*Calcolo* **2020**, *57*, 7, https://doi.org/10.1007/s10092-020-0357-9.