

## Research Article

# Polyurethane Application to Transform Screen-Printed Electrode for Rapid Identification of Histamine Isolated from Fish

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The toxicity of histamine has attracted numerous researchers to develop a method for histamine determination purposes. The Food and Drug Administration (FDA) unequivocally prohibits the consumption of histamine above  $50 \text{ mg}\cdot\text{kg}^{-1}$ . Thus, an innovation in histamine detection in fish has been developed in this research. The investigation of the histamine level in fish has been conducted by using an electrochemical sensor approach and producing a polymer via molecularly imprinted polymer (MIP) on a screen-printed electrode. The technique was validated by assessing the shifts in electron shifting using the cyclic voltammetry (CV) approach and electrochemical impedance spectroscopy (EIS), whereas differential pulse voltammetry (DPV) was applied to validate the sensor method. The instruments showed a linear response ranging from  $1\text{--}1000 \text{ nmol}\cdot\text{L}^{-1}$ , with a detection limit of MIP/SPE at  $1.765 \text{ nmol}\cdot\text{L}^{-1}$  and  $709 \text{ nmol}\cdot\text{L}^{-1}$  for the NIP/SPE, respectively. The sensing technique was employed to determine the histamine level in selected samples at room temperature ( $25^\circ\text{C}$ ). The outcomes of this study indicated that the validated chemical sensor allowed accurate and precise detection of fish samples and can be categorized as a simple approach. The instrument is inexpensive and suitable for on-site detection.

## 1. Introduction

The safety of food has become an imperative issue that must be handled properly owing to the ease with which food can be contaminated in the food chain system [1]. A statement has been reported by the World Health Organization (WHO) that one in ten people gets food poisoning and almost half a million die from food poisoning [2]. Even in Southeast Asia, particularly in Indonesia and Malaysia, food poisoning causes 22.8 million cases of diarrhea and 37,600 fatalities [3]. Furthermore, the food must be secured, and monitoring the food is imperative, both globally and in the Southeast Asia region [4]. Among various foods that have

been spread all over the world, aquatic foods have been related to food security issues owing to the massive consumption of aquatic food products in Southeast Asia [5, 6]. It is very common that physicochemical reactions can occur during the storage of food. The reactions can be influenced by several factors, such as heat, light, the packaging process, and moisture; furthermore, the addition of food adulterants can lead to the alteration of food content and cause health threats to humans [7].

Aquatic food products have a high nutritional value that is very useful for humans; however, compared to land animals, they contain more oils and fats, which cause several issues such as obesity, diabetes, and cardiovascular problems

[8]. Among all kinds of aquatic foods, fish is the best choice, according to nutrition experts; it also has low fat and high protein [9].

Nevertheless, the lucrativeness of fish may change into a problem owing to the decarboxylation process that occurs inside the fish body, producing biogenic amines. During the decarboxylation process, the amino acids in fish muscle become biogenic amines. The most popular biogenic amine, and a hot topic among researchers, is histamine. The consumption of histamine may lead to food toxicity, also known as histamine poisoning. Several effects may occur owing to the presence of histamine inside the bloodstream, and various symptoms occur, ranging from mild to even causing death [10, 11].

As a consequence, several countries have issued regulations for fish consumption that contain histamine. The United States of America (USA), through the Food and Drug Administration (FDA), only allowed  $50 \text{ mg}\cdot\text{kg}^{-1}$  of histamine, whereas the European Union (EU) has set the histamine level between 100 and  $200 \text{ mg}\cdot\text{kg}^{-1}$  [12]. Moreover, food poisoning from histamine is commonly related to several particular aquatic animals, such as sardine, mackerel, tuna, anchovy, and herring. Moreover, the development of a cheap and fast technique for histamine detection is imperative [13].

Numerous approaches have been performed to determine histamine, and most of them are chromatographic-based. However, there are several issues before using these methods, such as long-term analysis, complex instruments, and the need for derivatizing agents to derivatize histamine and increase the sensitivity of HPLC [14, 15], whereas the application of gas chromatography (GC) is restricted to laboratory facilities and complex preprocessing methods and usually requires a derivatization process, which is unsuitable for daily and on-site detection of histamine [16, 17]. The enzyme-linked immunosorbent assay (ELISA) offers a specific method for histamine detection, but this technique requires a specific environment to maintain detection precision, accuracy, and stability [18].

The colorimetric method becomes a better choice compared to the conventional methods, such as the straightforward technique, because it is fast and has attracted numerous researchers to study it [19–21]. Nevertheless, there are restrictions to this method, such as the accuracy being very low, the extinction coefficient being insufficient, and the color resolution being low, which cause inaccurate estimation [22, 23].

Furthermore, biosensors for histamine detection are an alternative approach, where this technique combines a bioreceptor with a transduction scheme. Generally, biosensors offer several advantages, such as rapid response, low cost, ease of operation, and high selectivity and accuracy [24, 25]. The bioreceptors generally employed for biosensors are antibody or enzyme materials, and they have satisfactory selectivity [26, 27]. However, they are also very expensive and have low stability caused by several factors such as pH, ionic content, humidity, and high temperature [28]. As a solution for receptor material, there is specific material such as molecularly imprinted material (MIP) that offers

several superiorities such as stability at high temperatures, robustness, high reproducibility, and on-site application [29]. Generally, MIP is a synthetic material produced by cross-linking monomers with polymerizing functions [30, 31]. Furthermore, the MIP properties propose satisfactory stability, easy production, a low budget, and robustness [32].

Electrochemical sensor approaches have a specific regulation that is related to the electrical parameters to build on the electrode surface. The specification of the sensor must be fulfilled to produce a satisfactory reproducible material and, consequently, obtain a satisfactory sensing instrument. Nevertheless, the process is not straightforward when the analyte experiences oxidation below its potential values [33], particularly histamine. Therefore, there are several MIP materials for histamine detection [34]. Thus, according to our findings, there is only one study that produces the MIP by electropolymerization [35].

This study conveys an electrochemical sensor for histamine detection on-site for the first time through the combination of MIP material with electropolymerized polyurethane (PU). PU is a unique polymer that has several advantages, such as good stability, being easily produced, being inexpensive, and having sufficient electrical properties that can be applied for histamine detection using the electrochemical sensor approach [36]. Furthermore, the MIP film was acquired by choosing the best states to develop a polyurethane-based imprinted film. The validated chemical sensor was validated and verified and then used to analyze histamine in selected samples.

## 2. Materials and Methods

**2.1. Instruments.** Electrochemical assessments were applied by a Metrohm Autolab Electrochemical Workstation provided by the Faculty of Science and Technology, Universiti Kebangsaan Malaysia. The SPE was a working electrode, while the reference and auxiliary electrodes were the Ag/AgCl with a double-junction system and platinum wire, respectively.

**2.2. Reagents.** All reagents were applied without purification. Histamine (His) ( $\geq 99\%$ ) was purchased from Sigma Aldrich Sdn. Bhd. Cadaverine (Cad), putrescine (Put), tyramine (Tyr), heptylamine (Hep), and spermidine (Spe) were bought at 99% purity and purchased from Sigma Aldrich Sdn. Bhd. Potassium hexacyanoferrate III ( $\text{K}_3 [\text{Fe}(\text{CN})_6]$ ) and potassium hexacyanoferrate II-3-hydrate ( $\text{K}_4 [\text{Fe}(\text{CN})_6] \cdot 3\text{H}_2\text{O}$ ) were obtained from Merck Sdn. Bhd. Sulfuric acid ( $\text{H}_2\text{SO}_4$ ),  $\geq 97\%$ , and lithium perchlorate were purchased from Sigma Aldrich, and polyurethane (PU) was produced by Munir et al. [36]. The phosphate buffer solution (PBS) was obtained by the combination of sodium dihydrogen phosphate ( $\text{Na}_2\text{H}_2\text{PO}_4$ ) and disodium hydrogen phosphate ( $\text{Na}_2\text{HPO}_4$ ), and both reagents were purchased from Merck, Sdn. Bhd. Ultrapure Milli-Q water was applied throughout.