High-accuracy Prediction of Meat Expiration Dates by Overcoming Nonlinearity Barriers for Microcantilever Biosensors

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Food waste is a major contributor to greenhouse gas emissions (GHG). It follows that animal-based food waste is responsible for more emissions than plant-based food waste. Waste can occur at various stages of the supply chain, and conservative shelf-life settings and subjective quality index measurements can lead to food waste, especially at the consumer level. The scientific spectroscopic and viable bacterial quantification methods currently used for measuring the quality and freshness and determining the shelf life are costly and time-consuming. Besides that, they are difficult to employ across the different stages of the supply chain. It follows that electronic nose (E-nose) technologies to measure freshness have been adopted, with a particular interest in the use of Piezoelectrically Driven Microcantilever sensors for the quantification of Cadaverine (1,5-Diaminopentane), a Volatile Organic compound whose concentration has been studied to increase with the level of spoilage. The response of these PD-MCs is, however, marred with nonlinearities, thereby affecting their accuracy and reliability.

This thesis proposes and demonstrates the use of PD-MC response models that account for the inherent and functional layer adsorption-dependent nonlinearities to enhance the accuracy of quantifying Cadaverine in a meat sample, thereby aiding accurate prediction of the shelf life (expiry date).

In this regard, this study begins by delving into the variables that impact the sensitivity of resonance frequency change in response to adsorbed mass, with a focus on the shape and size of the PD-MC. The unique design that is implemented involves a custom shape featuring a wide trapezoidal base housing the actuating piezoelectric layer, and a rectangular front where the functional layer is applied. The combination of these two shapes offers the best of both worlds in terms of advantages. An analysis is conducted to parameterize the response of the PD-MC, utilizing both experimental and analytical methods. The analysis includes free and forced vibration, utilizing the Euler-Bernoulli Beam theory and Hamilton's principle. The resulting data is then used to create an equivalent electrical circuit, which is simulated to determine the response to added masses. This information is then compared to experimentally obtained response data when the functionalized PD-MC is exposed to varying concentrations of Cadaverine. The parameterized response model is, however, found to be inadequate for low concentrations of Cadaverine where an increase in resonance frequency is observed.

After observing deficiencies in the parameterized response model, it was hypothesized that the increase in resonance frequency is caused by a chemical interaction between the binder and analyte, resulting in variations of the viscoelastic properties of the functional layer. To accurately quantify these variations, a combination of atomic force microscopy force curves and modeled fitting of the theoretical impedance response model of QCMs bearing a thin coating of the functional

layer was proposed and utilized. The study discovered that Young’s modulus of the functional layer increases with the increase of Cadaverine concentration at lower levels. As a result, the emergent viscoelastic changes effect dominates the changes effect of mass loading. The subsequent incorporation of the emergent adsorption-dependent response model produced a model that can accurately be used to quantify Cadaverine in a sample at lower concentrations. Furthermore, the Weibull reliability distribution was employed to calculate the ideal operating duration that would ensure accurate and dependable outcomes. The results indicate that a functionalized PD-MC can be employed with confidence for a duration of 152.69 minutes.

The validity of the models was confirmed by using them to measure Cadaverine levels in chilled tuna and Modified Atmosphere Packed (MAP) pork cutlets. This was done by comparing the results obtained through Liquid Chromatography with Tandem Mass Spectrometry (LC-MS/MS), Total Viable Count (TVC) of microbial flora, and Quality Index Measurement (QIM) based on human noses and visual assessment of appearance. When dealing with high concentrations of Cadaverine, the parameterized model, which accounts for the effects of mass loading was utilized. On the other hand, the adsorption-dependent nonlinear model was used for lower concentrations, where an increase in resonance frequency is observed due to the nonlinear dynamics of the functional layer. The response models showed trends that were in line with those obtained through LC-MS/MS measurements but demonstrated a nonlinear correlation across the number of storage days. It was determined that the differences in quantified Cadaverine levels were due to the adsorption of other biogenic amines in meat samples, leading to a suggestion that the Total Biogenic Amine Count is a more appropriate metric to use when comparing methods that quantify individual VOC concentrations.

To sum up, this study's findings showcase the promising application of PD-MC shape optimization and response models that consider the inherent and adsorption-dependent nonlinearities. This approach can effectively overcome the nonlinearity challenges in PD-MCs, leading to precise identification of VOC compounds as freshness indicators. Consequently, it enables accurate estimation of the shelf life concerning the production date.