

## COLORIMETRIC METHODS FOR FOOD FRESHNESS MONITORING: A REVIEW

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### ABSTRACT

#### Background

Food freshness monitoring has been a vital process in the food industry for ages. However, limitations are seen in the use of the usual laboratory and bulky equipment for chemical analysis. This led to the development of rapid, simple, and on-site colorimetric methods for food freshness monitoring based on visual color changes for quantitation and detection.

#### Scope and approach

In this review, the shift to simpler on-site determination is highlighted through the detection and quantitation of representative analytes signaling food freshness and food deterioration. The emphasis on the use of simple tools such as cameras and smartphones were highlighted through smart food packaging, colorimetric strips, and liquid reagents.

#### Key findings and conclusions

Colorimetric methods were seen to be a potential alternative to usual laboratory analysis counterparts. However, improvements in the key performance characteristics are needed to further advance this shift.

KEYWORDS: *colorimetry, food freshness, smart packaging, colorimetric films*

### INTRODUCTION

The shift from bulky, complicated, and costly chemical analysis to on-site, rapid, and simple chemical analysis and determination has been on the forefront of chemical research for quite a few years already (Wang et al., 2022). The preference for the latter has been already prevalent in various fields of science (Guo et al., 2020), specifically in the food safety (Chen et al., 2019) and manufacturing industry. Numerous methods have been developed to monitor food freshness and food deterioration (Liu et al., 2019), utilizing different chemical principles such as optical activity, gravimetric relationship, electrochemical methods, and one of the most common, colorimetric methods of analysis.

In colorimetric analysis, color parameters such as red, green and blue (RGB) and hue, saturation and value (HSV) are measured using cameras and other digital imaging hardwares to correlate such to analyte concentrations or pH, and ultimately to food freshness (Lee et al., 2019). Colorimetric analysis may be quantitative based on color-concentration correlations or qualitative based on visual color changes to signal chemical reactions or presence or absence of the analyte of interest such as ammonia and the like (Hashim et al., 2022).

This study aimed to review research articles on food freshness monitoring based on colorimetric methods. Specifically, this study aimed to 1) evaluate the colorimetric methods based on a) matrix, b) analyte, and c) sensing elements, d) performance characteristics

utilized, and e) real samples used, and 2) determine the a) hardware specifications, b) color parameters used, and c) products developed.

### METHODS

This review paper utilized a structured keyword search on colorimetric methods for food freshness monitoring using keywords a) food freshness and b) colorimetric, considering articles written in the 2019-2022 timeframe using Science direct as search engine. Initially, 125 articles were considered but were trimmed down to 40 articles, focusing on a) analyte, b) matrix, c) instrumentation, d) sensing element, and e) application.

#### 3.1 Matrix and Sample types

Samples analyzed in the various research articles included solid and liquid samples containing varying analytes of interest.

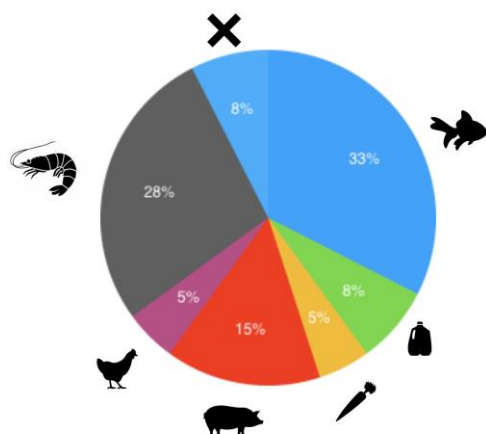


Figure 1: Matrix distribution

Figure 1 reveals that majority of the matrices used in the articles are seafoods, with 28% utilizing shrimp as primary matrix for analysis. This result may be attributed to the fact that marine animals produce easily detectable amines during their deterioration process (Nativ et al., 2021). Amines were also seen as the primary analyte used in majority of the reviewed articles, as amines are important markers for food freshness and safety (Zhu et al., 2020).

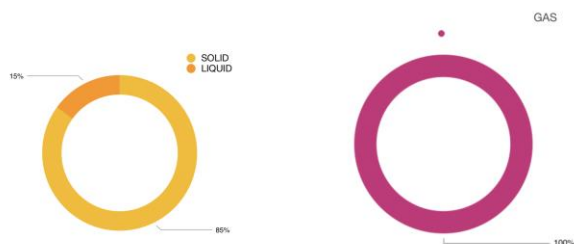


Figure 2: Matrix distribution(left) and Analyte type(right) utilized

Figure 2 shows the huge preference in using solid matrices, accounting to 85% of the reviewed articles. Sample solid matrices included fresh or frozen seafoods (61%) and vegetables (5%) while liquid matrices included milk as an example (8). It is worth noting that 100% of the analytes examined in the various articles were in the gaseous form, usually belonging to the amine functional groups with some exceptions like carbon dioxide.

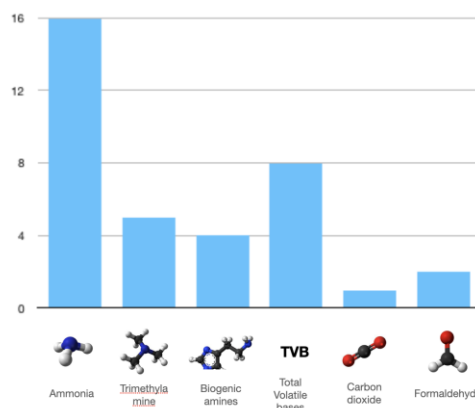


Figure 3: Analytes used in the research articles

### 3.2 Analyte types

Only carbon dioxide and formaldehyde (Wongniramaikul et al., 2018; Wu et al., 2021) were the analytes not belonging to the amine functional groups. The huge preference for amines as analytes of interest rests on the fact that these compounds are formed during the degradation of seafood and other organisms (Nativ et al., 2021).

High concentrations of the analytes usually signal poor freshness or advanced rate of deterioration in chosen food samples (Zhu et al., 2020).

### 3.3 Performance characteristics

In order to ensure that the methods developed are as accurate and precise as their corresponding laboratory counterparts, performance characteristics were also analyzed. It is worth noting, however, that only a few were able to cover most of the performance characteristics such as accuracy and precision, limit of detection, reproducibility and repeatability, and other analogous and relevant performance characteristics. The most commonly evaluated performance characteristic was the working range. In this review, majority reported their working ranges as a pH range rather than the usual analyte concentration range. A working range of pH2-pH12 was dominantly reported in majority of the reviewed articles. This means that the colorimetric methods have a wide range of applicability covering almost the full pH scale of pH1-pH14. As previously discussed, since the analytes of interest were majority belonging to the amine functional group, it is easy to correlate the pH to food freshness because of the pH changes caused by varying amine concentrations.

### 3.4 Real samples

Based on figure 1, only 8% of the reviewed articles did not utilize real samples, and only relied on standard solutions/mixtures to proceed with the experiment. 92% utilized real samples, including fresh or frozen samples in the process.

### 3.5 Sensing elements

One of the most essential component in colorimetric analysis is the sensing element. This ensures that the proper color change is elicited in the process, thus providing the basis to carry out the preferred chemical analysis. Colorimetric methods have been utilized in the food safety field because of its convenience and use of nanomaterials such as nanoparticles, nanorods, and other sensing materials (Qi et al., 2020).

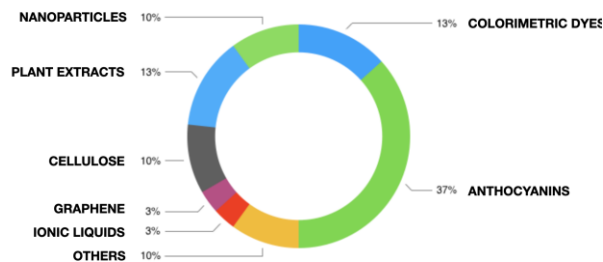


Figure 4: Sensing elements utilized

Figure 4 presents an overwhelming preference to anthocyanins. Generally, anthocyanins are natural compounds which are pH-sensitive and pH-dependent (Kang et al., 2020). This means that they can easily change color when the pH of the solution or environment changes (Wu et al., 2020). Thus, they are seen to be excellent color changing indicators in 37% of the research articles reviewed. A handful of the reviewed articles utilized colorimetric dyes (10%), nanoparticles (10%), and other analogous sensing elements such as graphene, ionic liquids, and the like (Vovanska et al., 2022). Over-all, the sensing element must be able to elicit the proper color change needed to proceed with the colorimetric analysis (Qi et al., 2020).

Table 1 shows some representative works of the reviewed articles showing relevant information such as analyte, sensing element, and application.

Table 1: Representative studies

Analyte	Sensing element	Application	Reference
Trimethylamine	Anthocyanin	Food packaging	Vovanska et al., 2021
Total volatile bases	Anthocyanin	Colorimetric film	Zeng et al., 2019
Biogenic amines	Graphene oxide	Food packaging	Siripongprada et al., 2020
Ammonia	Ionic liquids	Colorimetric films	Ballester-Caudet et al., 2021
Formaldehyde	Gold nanoprisms/Tollen's reagent	Liquid reagent	Qi et al., 2020

In general, various applications such colorimetric films, food packagings, smart sensors, and liquid reagents were utilized in the majority of the reviewed literature.

### 4.1 Hardware specifications and requirements

Hardware specifications and requirements play an essential role in data acquisition and processing. This guarantees that the acquired color parameters are precise and accurate.

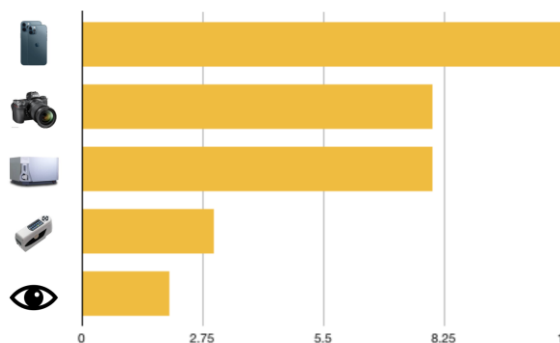


Figure 5: Equipment used to acquire data

True to the objective of shifting to on-site and rapid colorimetric chemical analysis, smartphones and Digital single lens reflex cameras (DLSR) were utilized as the leading equipment (Qi et al., 2020), followed by the reliable and standard spectrophotometers used in the laboratory. It is also worth noting that some studies utilized the use of the naked eye for visual color change interpretation.

#### 4.2 Image resolution and analysis

Image analysis applications such as image J, adobe photoshop, and other relevant commercial color applications were utilized. This further justifies the convenience of using colorimetric analysis in chemical analysis as all these applications are easily accessible and can be downloaded or bought from providers in smartphones or computers.

#### 4.3 Color parameters utilized

Basic colorimetric parameters such as RGB and HSV were utilized in the study. Qualitative and quantitative analysis related to color changes were performed as these were basis for successful determination and analysis (Sutthasupa, 2021).

#### 4.4 Products developed

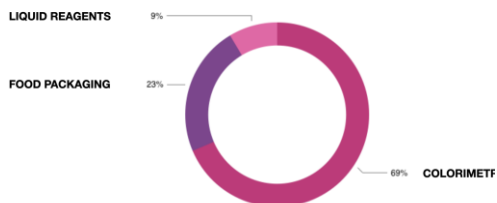


Figure 6: Products developed

The products developed in the study depended on the application and matrices utilized. Three major products were seen to be developed, these were liquid reagents, food and smart packagings, and colorimetric films. All were geared toward the determination of freshness levels of the food sample of interest.

#### Conclusion

The following conclusions are hereby derived:

1. Colorimetric methods of analysis provide a rapid, cost-efficient means of chemical analysis. This can

greatly improve and revolutionize the food freshness monitoring in the food safety field. Interestingly, majority of the colorimetric methods reviewed in the recent years of 2019 with majority heavily relying in pH changes eliciting the colorimetric responses in films and food packagings with the use of sensing elements such as anthocyanins;

2. Majority utilized amine functional groups as primary analyte of analysis;
3. No direct comparison between standard methods (laboratory counterparts or other on-site methods) were seen and done; and
4. Due to the simple and uncomplicated nature of the experiments, a sharp rise in the preference of colorimetric methods is evident.

#### Summary and future insights

Moving forward, the following improvements can be explored and applied to further the shift to colorimetric methods for food freshness monitoring:

1. Key performance characteristics must be explored and evaluated;
2. Performance against standard methods must be explored;
3. Working ranges must be expressed not only in pH but also in terms of concentrations of the chosen analytes; and
4. Creation of applications and softwares may be looked into after standardization of methods.

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