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A Review of the Different Sensors for the Detection of Rice Rancidity Indicators

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Rice loss occurs in various supply chain stages ranging from harvesting to consumption. Within this chain, rice is handled and stored by farmers, distributors, and vendors, not all of whom would likely have access to sophisticated storage facilities and management processes. Non-ideal storage conditions can lead to rancidity – the spoilage of food to such an extent that it becomes undesirable or even unsafe for consumption. To reduce the wastage of rice during warehousing and storage, sensors that will monitor the onset of rancidity are necessary to mitigate losses associated with spoilage. This work summarizes the different sensors that have been developed to detect the onset of rancidity in rice. Various types are discussed, as well as the corresponding parameters that affect the sensor response, detection limit, and sensitivity. Polymer-based sensors are superior to transition metal-based sensors with a polymer complex that can detect up to 0.05 ppm of hexanal and has the best performance reported in the literature. Recent developments are elucidated as to their application in improving the monitoring of rice conditions during warehousing, storage, and transportation.

1. Introduction

Rice is considered one of the staple foods produced in the world. It is considered a major food source in Asia with more than 90 % of the worldwide production. Rice is an essential source of energy, minerals, fiber, vitamin E, phenolic acids, anthocyanin and proanthocyanin, flavonoids, phytosterol, carotenoids, phytic acids, tannins, and γ-oryzanol required for the proper functioning of the human body. Rice biomolecules contain anticancer, anti-inflammatory, antioxidant, antidiabetic, antibacterial, antiarthritic, and hypoglycemic activity, which have effective health benefits (Sen et al., 2020).

The production and consumption of rice is an important activity for more than half of the world's population. It is estimated that the production of rice will rise from 58 to 567 Mt in 2030 (Myszkowska-Ryciak et al., 2022). However, anthropogenic activities that cause drought, the toxicity of irrigated lands, rising sea levels, or flooding of low-lying areas constrain rice production, which could affect 30 % of the 700 million people in Asia who rely on the crop. The International Rice Research Institute promotes research on new rice varieties that can withstand environmental stresses such as drought, nutrient deficiency, high salinity, iron toxicity, sodicity, submergence, and low temperatures. The design of new rice varieties with high tolerance to environmental factors would allow a high yield of the crop even in the most inhospitable location. Aside from environmental factors affecting the production of rice, spoilage during transportation and storage poses a significant threat to food security (Que et al., 2021).

Rice is grown and harvested over a limited period yet consumed throughout the year. The processing and storage after harvesting of grains significantly affect their quality. It is estimated that up to 50 % - 60 % of cereal grains are lost predominantly to improper practices and lack of awareness of scientific and technological tools. The losses can be brought down to 1 % - 2 % with proper scientific know-how (Darmawan et al., 2018). Aulakh et al. (2013) provided an estimation framework for post-harvest food losses and described that maximum losses occur during the storage stage. Much of the losses associated with rice grain storage in warehouses are due to moisture content and fluctuating temperature. Fresh harvest contributes to heat and moisture due to respiration, which can further exacerbate spoilage conditions. The heat and moisture generated can contribute to mycotoxin development, mold growth, and fungal and bacterial contamination, which are causes of rice rancidity (Muller et

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al., 2022). Fortunately, there are various ways available to prevent or delay spoilage of rice grain during transportation and storage. Simple steps can be carried out, such as monitoring of temperature and moisture content and proper aeration to reduce losses in warehouses. In this aspect, continuous monitoring technologies using different sensors could provide valuable information about the condition of the grains in real-time. Any changes associated with fluctuation in environmental conditions can be addressed early on before the onset of detrimental growth of microorganisms that can cause rice rancidity.

Specific chemical signatures can be used as indicators of rice rancidity. Aldehydes have been detected during the onset of rancidity. Hexanal has been a target molecule, and the onset of rancidity has been ascribed to the presence of this volatile organic molecule (Kishimoto, 2021). Polymer-based sensing elements have been synthesized and demonstrated to have better performance than transition-metal-based sensors, as reported by Wang et al. (2020), with a detection limit of 0.05 ppm. Despite the popularity of rice as a staple food for much of the world, research on sensor technology for monitoring rice quality is scarce, and a gold standard for sensors has not yet been established. Recent developments in rice sensors have yet to be reported in the literature. This paper is conceptualized to provide status on the state and future prospects of rancidity sensors for the detection of the onset of rice spoilage.

2. Rice Rancidity

Rice products sold in the marketplace require proper handling and storage procedures to avoid undesirable off-flavors, which several factors could cause. The usual cause of food spoilage is microbial; however, quality deterioration could also be due to chemical changes brought by enzymatic reaction, hydrolytic reaction, or oxidative alteration of the chemical and physical properties of the rice product. The term associated with off-flavor and undesirable odor associated with food or grain deterioration in quality is called rancidity.

Rancidity can be classified based on two basic types: oxidative and hydrolytic. Oxidative degradation of rice quality is associated with oxidation of the lipid due to the presence of oxygen gas and free radicals. On the other hand, hydrolytic degradation of rice quality is associated with moisture content, which causes unsaturated lipids to be cleaved into low molecular weight aldehydes and ketones.

2.1 Oxidative Rancidity

Oxidative rancidity is caused by oxidative deterioration of unsaturated fatty acids due to the presence of oxygen. Light and heat promote this reaction, resulting in the generation of low-molecular-weight aldehydes and ketones. When lipids are oxidized through a complex multi-step process, volatile and non-volatile compounds are emitted, which causes the undesirable odor and flavor associated with food deterioration (Shahidi, 2015).

Oxidation of lipids proceeds through the following stages: initiation, propagation, and termination. Enzymes or other catalysts initiate the breakdown of lipid molecules to produce reactive intermediates or free radicals. These free radicals, which contain unpaired electrons, are very reactive and cause the oxidation of other compounds in the food/grain. Hydroperoxides and other oxidation products, such as aldehydes, ketones, and organic acids, could be produced when these radicals react with lipid molecules. Aldehydes such as hexanal are volatile compounds considered to have a low odor threshold, and their concentrations increase with storage. These compounds are the indicators of the rancidity of food/grain.

2.2 Hydrolytic Rancidity

Hydrolytic rancidity occurs when triglycerides are hydrolyzed to form free fatty acids. The process either requires a catalyst or not. Rancidity can vary from barely noticeable, indicated by loss of freshness, to strong odor emanating from the chemical reactions.

2.3 Factors Affecting Rancidity and Rancidity Indicators

There are several factors that affect the oxidation of lipids, such as sunlight, high temperature, humidity, exposed surface, nitrogenous organic material, air, and traces of metals (Muller et al., 2022). To prevent or minimize the rate of lipid oxidation, the grain or food product must be stored in a place shielded from direct sunlight, with low exposure to oxidizing agents, low temperature, and free of metal catalysts.

Several compounds are produced from the oxidative and hydrolytic degradation of fatty acids. During the process of oxidative reaction of lipids, peroxides are produced. This forms the basis of the peroxide value (PV) method of rancidity testing. However, further reactions occur during rancidification and the ultimate product of hydrolytic and oxidative rancidification are ketones and aldehydes (Shahidi, 2015).

3. Analysis of Rancidity Indicators

Rancidity in rice represents a significant quality concern, affecting both its sensory characteristics and nutritional value. It is essential to understand and accurately detect indicators of rancidity to ensure the quality of rice during storage and distribution. This section explores various analytical methods utilized to identify and quantify rancidity indicators in rice. Spectroscopic, chemical, colorimetric, and chromatographic techniques are explored, each providing unique insights and benefits for assessing rice quality. By employing these methods, it becomes possible to monitor and mitigate the effects of rancidity, extending the shelf life and preserving the nutritional integrity of rice.

3.1 Spectroscopic Methods

Several spectroscopic techniques have proven effective in analyzing rancidity indicators in rice, leveraging the ability of these methods to provide detailed chemical profiles. Ribeiro et al. (2020) demonstrated the utility of laser-induced breakdown spectroscopy (LIBS) and Fourier transform infrared spectroscopy (FTIR) in differentiating rice varieties based on their chemical composition. These methods enable rapid, non-destructive analysis, offering insights into the presence of rancidity indicators such as free fatty acids and peroxides. Joshi et al. (2015) highlighted the potential of hyperspectral imaging and vibrational spectroscopy for non-invasive assessment of rice quality, particularly in detecting changes in starch and protein content during storage. Such changes are critical indicators of the onset of rancidity and can influence the texture and taste of rice. Additionally, Natsuga and Kawamura (2006) supported the use of visible and near-infrared reflectance spectroscopy to determine the physicochemical properties of rice, such as moisture content and appearance. Maintaining optimal moisture levels is essential in preventing rancidity, as excess moisture can accelerate the degradation of oils within the rice grains. Spectroscopic methods have been widely employed in practical applications by rice producers and quality control laboratories. The FTIR spectroscopy is routinely used in industry to monitor the oxidative stability of rice bran oil, a critical factor in extending the shelf-life of rice products. This method allows for the quick detection of lipid oxidation products for early indicators of rancidity.

3.2 Chemical Methods

Various studies have explored the application of different chemical indicators to assess rancidity in rice, focusing on the precision and specificity these methods offer. Kim et al. (2014) found that specific heat treatments, such as autoclaving, can effectively retard the formation of free fatty acids, which are key indicators of rancidity. By applying controlled heat treatments, it is possible to prolong the shelf-life of rice without compromising its nutritional value. Ribeiro et al. (2020) utilized LIBS and FTIR to detect subtle chemical variations in different rice types, focusing on protein, fatty acids, and magnesium. These elements play significant roles in the nutritional quality of rice, and their alteration can signal the early stages of rancidity. These studies collectively underscore the importance of chemical analysis in providing detailed, quantitative assessments of rancidity and other quality indicators in rice.

3.3 Colorimetric (Optical) Methods

Colorimetric analysis methods have been widely applied to assess rancidity indicators in rice, leveraging the visual changes that occur as rice deteriorates. Jinorose et al. (2010) employed image processing techniques to evaluate rice color by focusing on developing computer-vision algorithms and evaluation criteria. These techniques allow for the automation of quality control processes, making it easier to monitor large quantities of rice efficiently. Kishimoto (2021) adopted a different approach, using a colorimetric sensor array combined with volatile organic compounds to distinguish between fresh and aged rice. This method provides a non-invasive means to detect spoilage through the analysis of emitted volatiles, which are indicative of rancidity. Joshi et al. (2015) established a correlation between color parameters and sensory characteristics of rice, offering a potential method for predicting eating quality. By linking visual characteristics with sensory properties, it becomes possible to assess rice quality without the need for extensive chemical tests. These studies collectively demonstrate the potential of colorimetric analysis in providing rapid, cost-effective assessments of rancidity indicators in rice.

3.4 Chromatographic Methods

Several chromatographic methods have been explored for analyzing rancidity indicators in rice, providing high-resolution separation and identification of complex mixtures. Shokrzadeh et al. (2007) highlighted the potential of high-performance liquid chromatography (HPLC) in identifying rice varieties and analyzing endosperm proteins. HPLC allows for the precise quantification of compounds associated with rancidity, such as free fatty acids and lipid oxidation products. This is further supported by Kim et al. (2014), who found that various heat treatments can affect the rancidity and bioactive compounds in rice bran. By understanding how processing conditions impact rice composition, it is possible to develop methods to mitigate rancidity while preserving beneficial nutrients. These studies collectively demonstrate the potential of chromatographic methods in providing detailed, accurate analyses of rancidity indicators in rice. Chromatographic methods, combined with other analytical techniques, offer a comprehensive approach to ensuring the quality and safety of rice throughout its shelf-life.

4. Polymer-Based Sensors and Electrochemical Sensors

Polymer-based sensors are more effective in the sensing of hexanal molecules compared to inorganic oxide sensors. The mode of interaction is due to the reversible interaction of the specific functional groups in the polymer to the target analyte. The inorganic electrochemical sensors, on the other hand, can be divided into three major types: (1) potentiometric sensor, where the sensing element measures the voltage between the reference and working electrode; (2) amperometric sensor, where detection is based on the amount of current generated from the oxidation or reduction of the analyte species, and (3) impedimetric/chemo resistive sensor, where both resistive and capacitive changes in the system are measured and reported as impedance.

In this paper, we focused on the detection of aldehyde compounds released from the degradation of rice quality. Hexanal is one particular compound that has been detected as a byproduct of the early stage of lipid oxidation for food products (Shahidi, 2001). Sensors for low molecular weight aldehydes were also discussed due to the similar functional group with hexanal.

4.1 Polymer-Based Sensors

Polymer-based composite materials have been proven to be effective in selectively detecting hexanal as a rancidity indicator, as shown in Table 1. Shantini et al. (2016) developed a chitosan biopolymer using electrochemical deposition. It was found to have good selectivity and response towards hexanal at room temperature. The electrodeposition technique was also applied by the same group in the development of chitosan/polyvinyl alcohol (PVA) on gold-patterned electrodes for hexanal detection. The group of Wang et al. (2020) developed a polydopamine-polyethyleneimine (PDA-PEI) copolymer film for the detection at the ppb level. The sensor was found to have effective sensing output due to reversible hydrogen bond formation with hexanal. In addition, the moisture resistance of the material can be improved by the addition of n-octadecylsiloxane onto the surface. In the sensor designed by Chen et al. (2019), hexanal could selectively bind with molecularly imprinted polymer (MIP) composite. It was found to have a low activity compared to other volatile compounds such as ethanol, acetone, acetic acid, and ammonia. Hexanal is also used as a biomarker for the diagnosis of lung cancer in the studies of Guo et al. (2022). The results of their studies could also be applied to the detection of hexanal in food due to similar target molecules. In the study of Mousazadeh et al. (2022), they were able to design a sensor that can detect up to 1.1 ppm concentration of the target molecule. In all these developed sensors, the mode of detection is based on the reversible interaction of functional groups on the polymer molecule with the target hexanal compound.

Table 1: Polymer-based sensor for hexanal as rancidity indicator

Active Material	Aldehyde	Detection Limit (ppm)	Synthesis Technique	Reference
Chitosan/PVA	hexanal	10 - 30	Electrodeposition	Shantini et al., 2016
n-octadecylsiloxane-functionalized PDA-PEI	hexanal	0.050	Co- and self-polymerization	Wang L. et al., 2020
Hydrophobic polymer composite	hexanal	14 - 27	MIP	Chen et al., 2019
Au nanoparticles/MIP	hexanal	1.1	MIP	Mousazadeh et al., 2022

4.2 Inorganic Electrochemical Sensors

Inorganic-based sensors are effective in the detection of low-molecular-weight aldehydes, as shown in Table 2. Many types of metal oxide sensors have been synthesized. An exhaustive treatment for food application was discussed by Berna (2010). Various shapes, sizes, and porosities have been prepared using different preparation techniques. Improvements have been introduced, ranging from synthesis method, addition of dopants, combination with other metal oxides, improvement of designs, and sensing elements.

In the study of Li et al. (2019), a wearable IoT aldehyde sensor was developed that can detect formaldehyde levels up to 30 ppb. It was based on a fuel cell sensor connected to electronic components for continuous formaldehyde monitoring for up to seven days. Integration of aldehyde sensors to IoT would be very effective for continuous monitoring of rice quality to avoid wastage during transportation and storage.

Table 2: Transition metal oxide sensors for aldehydes

Active Material	aldehyde	Response (s)	Detection Limit (ppm)	Synthesis Technique	Reference
InO_3	acetaldehyde	-	1 - 100	Microwave hydrothermal	(Chava et al., 2019)
CuO	acetaldehyde	9293	10	Chemical synthesis	(Patil et al., 2019)
α-Mn_2O_3	acetaldehyde	9	5 - 75	Spray pyrolysis	(Srinath et al., 2015)
Co-doped InO_3	formaldehyde	23.2	10	hydrothermal	(Wang et al., 2018)
F-doped ZnO	acetaldehyde	4.8	100	electrodeposition	(Gunasekaran et al., 2018)
Ni-doped ZnO	acetaldehyde	2.59	10	Spray pyrolysis	(Mani and Rayappan, 2016)

5. Challenges and Future Directions

The successful detection of the onset of rice spoilage can save enormous savings and economic benefits to farmers, traders, rice importers, consumers, and governments. As a result, it can minimize wastage due to rice grain spoilage and can provide a sustainable path to food security. However, technical challenges associated with the design and fabrication of electrochemical sensors must be overcome. Interference of other chemical compounds present in the rice matrix can result in false positive and negative results. This can be avoided by improving the specificity and selectivity of electrodes. Currently, practical applications are limited by the design of highly active electrodes that target the molecules. Due to the variety of compounds emitted during rice degradation, it is hard to design an electrode that can capture all chemical signatures and, at the same time, be agnostic to interfering molecules such as ethanol or moisture. The long-term stability, durability, and robustness of sensors under various environmental conditions of heat, moisture, sunlight, and other factors are major considerations in reducing economic costs. There is also a necessity for standardized protocols for sensor calibration, data interpretation, and validation.

Future trends in the rice rancidity sensor include integration with the Internet of Things (IoT). This would allow faster data acquisition and interpretation, real-time monitoring, and data analytics for efficient management and logistics. Miniaturization of electrode sensors would allow easy handling for field application and deployment. Multi-analyte detection of the product of rice spoilage is another important characteristic that must be developed for future design of rice sensor arrays.

6. Conclusions

Sustainable agricultural practices coupled with proper monitoring and storage can reduce CO_2 emissions in the agricultural sector. Spectroscopic, chemical, and chromatographic techniques are highly accurate but require trained personnel and expensive instrumentation for continuous rice quality monitoring. Low-cost electrochemical and polymer-based sensors provide alternative solutions. A polymer complex that can detect up to 0.05 ppm of hexanal has been reported to have the best detection level. It is very promising, but a lot of work is still required for actual deployment. The selectivity requires improvement to avoid false positives from interfering substances, and sensitivity towards the target molecule can be further enhanced (i.e., using highly conductive and high-surface-area substrates).

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