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Intelligent pH Sensing Films in Food Packaging

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Recent rise in the awareness in the safe and changing consumer's attitude have bought many innovations in packaging technology. Monitoring the freshness of food products is essential during their manufacturing or handling because most commercially available foods are perishable in nature. Moreover, their organoleptic/nutritional qualities depend on changes in their environmental or physiological states, thereby causing spoilage and a possible food safety issue so packaging plays a main role in the food industries. Further, humans always prefer to consume foods that are generally esthetically pleasing in nature. Packaging plays an essential role in maintaining food quality, food safety and extending the shelf life of food in the food industry.

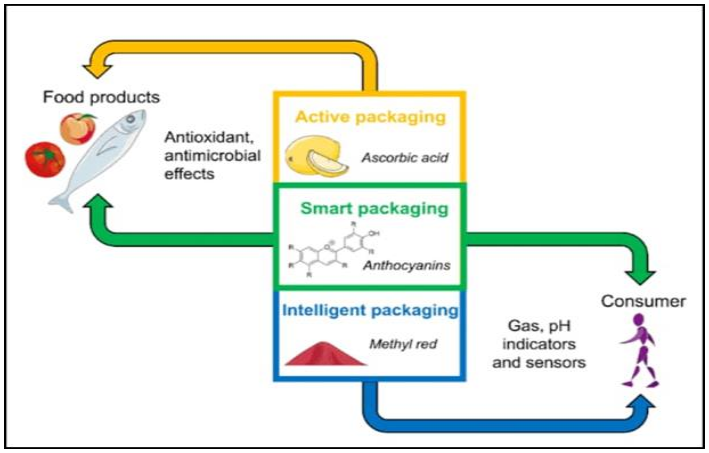

Fig. 1 Intelligent packaging

There is a new platform and are best taken out for the consumer acceptability in packaging. Intelligent packaging means monitor the status and environment of the food commodity and gives the information to the consumers about food safety and quality. It is an innovative approach that has been implemented to monitor various condition changes in the packaged products, including food quality (freshness or spoilage), microbial activity (generation of organic acids, CO_2, volatile nitrogenous substances, or sulphur derivatives) or other properties (transport and storage history that affect food quality). Moreover, this novel packaging strategy allows a preferential site that provides the necessary information regarding the delivery or preservation conditions of products and facilitates safety assessment, quality assurance and customer satisfaction. Intelligent packaging can sense, monitor, record, trace and provide information about the shelf-life, safety and quality of food.

Intelligent packaging system

Intelligent packaging system consists of indicators, sensors and Data carriers.

Indicators:

A. **Time-temperature indicators**: Time-temperature indicators (TTIs) are typically small self-adhesive labels attached onto shipping containers or individual consumer packages. These labels provide visual indications of temperature history during distribution and storage, which is particularly useful for warning of temperature abuse for chilled or frozen food products. They are also used as "freshness indicators" for estimating the remaining shelf life of perishable products.

B. **Oxygen indicators**: Oxygen indicators are mainly used for food packaging applications, because oxygen in air can cause oxidative rancidity, colour change, and microbial spoilage. A number of oxygen indicators are designed to show colour changes due to leaking or tampered packages.

C. **Freshness/ pH indicators**: Freshness indicators are direct food quality indicators, since they provide information on microbiological or chemical changes responsible for food spoilage. Freshness indicators for meat products, for example, may be based on biogenic or hydrogen sulfide (H_2S, released during meat spoilage). In seafood, the total volatile basic nitrogen content (volatile amines), primarily composed of dimethylamine (DMA), trimethylamine (TMA) and ammonia, is frequently used to assess microbial degradation. The indicators may be based on the

detection of volatiles in the headspace of food packaging.

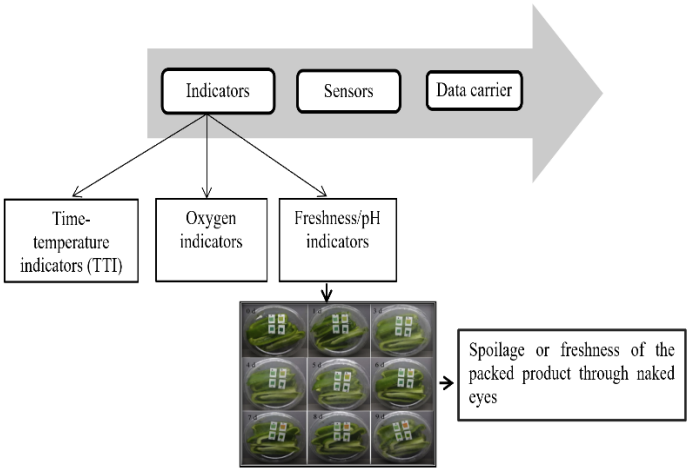

Fig. 2 Intelligent packaging system

Intelligent pH indicator/sensing films

Intelligent pH sensing films are essentially a pH-sensing dye embedded into a polymer matrix support. The indicator is then inserted into a primary or secondary packaging material where it provides real-time visual colorimetric response to the condition/state of the packaged food. Packaging with pH indicators films usually consists of two parts,

a. Solid base/ carrier material: Chitin, Cellulose and gelatin

b. Natural pigments/ dyes: Sensitive to the pH change that makes up the colorimetric sensor.

The natural pigments/dyes are extracted from various sources of fruits and vegetables. Intelligent pH sensing films have the ability to distinguish fresh, medium fresh and spoiled food typically by the naked eyes. Colour response, safety of the intelligent pH indicator depends on the type and content of the indicator dye, as well as the matrix support.

Chemical substances such as cresol red, bromophenol blue, bromocresol green, and bromocresol purple were applied as pH sensing dyes. Application of these synthetic dyes as intelligent pH indicators suffer from one major limitation: safety concerns due to potential toxicity. Hence chemical substances are replaced by natural pigments. Natural pigments such as anthocyanins are very sensitive as pH indicator dyes; their practical application is impaired by rapid colour fading due to oxidative degradation.

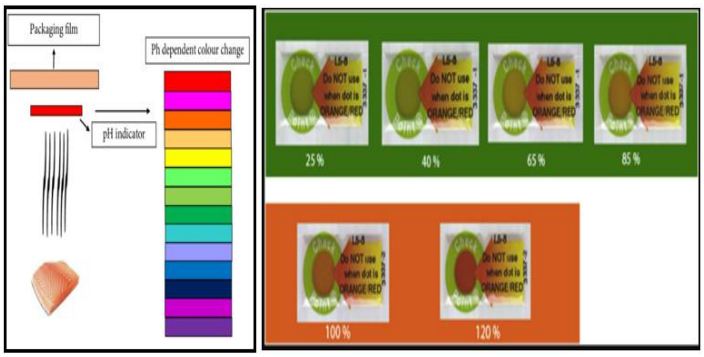

Fig. 3 pH-dependent colour change of food material

Preparation of Intelligent pH Indicator Films

For the preparation of film, film forming materials are very important hence to protect the environment from the hazard biopolymers are used as a carrier material such as, chitosan, cellulose and gelatin etc. For sensitive to the pH change that makes up the colorimetric sensor for this natural pigment *i.e.*, anthocyanin rich fruits or vegetables extracts are used. Initially extract should be prepared to check the variation at different buffer solution and also to check the pH range because different anthocyanins have different pH ranges. Example: Anthocyanin rich broken rice berry was pulverized for 2 min using grinder sieved through an 80-mesh sieve to obtain broken Riceberry powder (particle size < 180 µm). Broken Riceberry powder was (80% v/v) containing acetic acid (0.25% v/v) at 60°C stirring for 2 hrs. The supernatant was separated centrifuging at 4500 rpm for 10 min. The supernatant was decanted and filtered through Whatman Number 1 filter paper. Ethanol in the filtrate was evaporated under vacuum at 40 °C yielding a concentrated extract. The extract was left overnight at 4°C and then lyophilized to obtain broken Riceberry phenolic extract (RPE) powder. The RPE powder was stored at –20° C.

Chitosan solution (1% w/v) was prepared by solubilizing in acetic acid (1% v/v). Films were prepared by adding different amounts of RPE powder (0, 1, 2 and 3 w% on chitosan basis) to 58 mL of chitosan solution to give CHI, CHI-RPE-1, CHI-RPE-2, and CHI-RPE-3 solutions, respectively. Glycerol (0.38 mL) was added as plasticizer. The film solutions were poured on casting trays (13 × 10 cm) and dried at 37 ◦C for 36 h. The films were peeled off, followed by

conditioning at 25 ◦C in a desiccator with 50 % relative humidity for at least 72 h before testing.

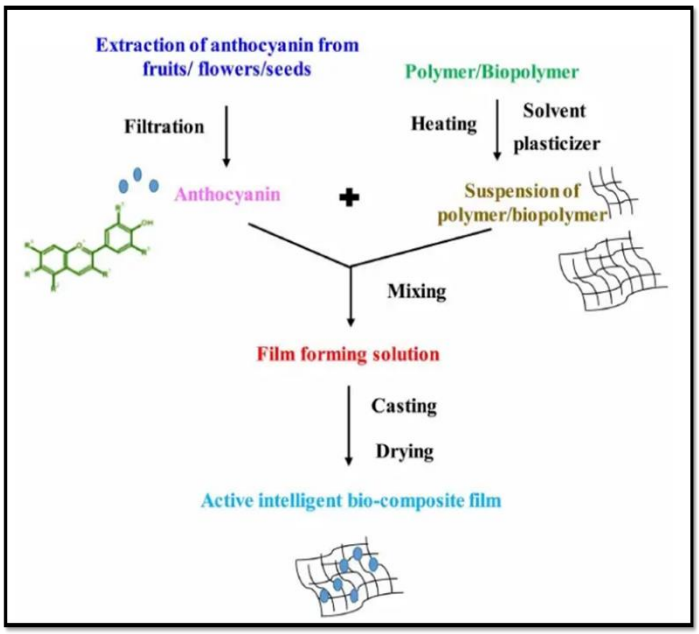

Fig. 4 Preparation of intelligent pH sensing film

Fabrication methods

Different fabrication methods are used for preparation of pH sensing films such as Nanoparticles immobilization process, Electrospinning and Casting.

a. **Nanoparticles immobilization process:** Naturally found colorimetric pH indicators are immobilized with various nanoparticles or nano polymers to enhance their colour stability and increase their ability to evaluate the spoilage stage caused by even slight colour variation.

b. **Electrospinning:** Electrospinning is a technique used for the fabrication of submicron-sized polymer nanofibers. In this method, different macromolecules are uniformly electrospun to form ultrafine fibers in the order of several nanometres upon the application of intensive electric fields. In this process, the electrically charged polymer jets from a high-voltage supplier are accumulated on the collector surface using a syringe needle.

c. **Casting:** Casting is a fabrication technique that is widely used to produce various fibrous nano-composite films for application in pH-sensitive indicators systems.

Colour changing mechanism of anthocyanin in response to various pH

pH sensitive indicator films change colours in response to the changes in the pH of perishable foods under varying storage conditions. This allows consumers to easily monitor the quality (fresh or spoiled) of their products without opening the package. There are several causes that may result in the pH change of food. Microbial decomposition of carbohydrates, proteins or fats may cause the production of various volatile, basic nitrogenous compounds, such as ammonia, trimethylamine and dimethylamine which ultimately increase the level of pH in the headspace of packaged products. Colour changes in the anthocyanin-based indicators films can be primarily attributed to the changes in the chemical structure of the anthocyanin in the film at varying pH in a mild or intensely acidic (pH 4 - 5) environment, the red flavylium cationic state of anthocyanin are converted to gray carbinol pseudo-bases by deprotonation which further turn to quinoid anhydrous bases at neutral pH (pH 6 - 8), exhibiting violet color. Similarly, in highly alkaline environment the indicator is converted to yellow chalcones.

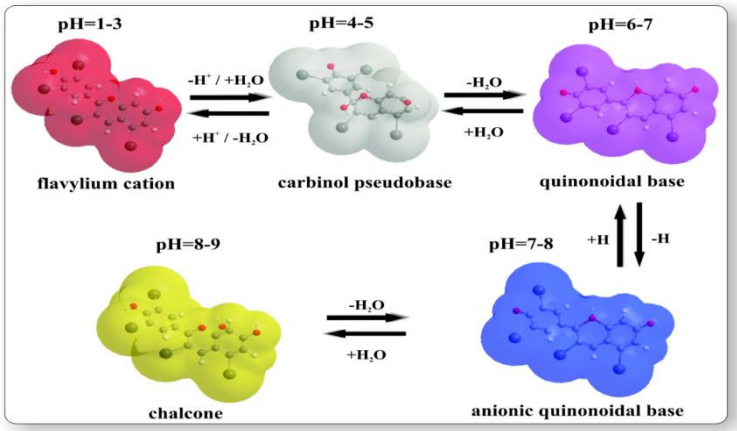

Fig. 5 Colour changing mechanism of anthocyanin in response to various pH

pH range for intelligent pH sensing films

pH-dependent colour changing properties of anthocyanins were used to develop indicator films for monitoring the pH change of packaged foods in real-time. The different anthocyanins are having different working pH ranges due to the varying storage conditions.

Triple layered freshness indicator, preventing migration from sensing element for food quality check

Intelligent pH sensing films are placed inside the packaging material in connect with the food or on the packaging material. Packaging material used is LDPE (Low density polyethylene). Film helps to indicate the spoil and freshness where the label will be placed on the top of the packaging materials to know the changes through naked eyes.

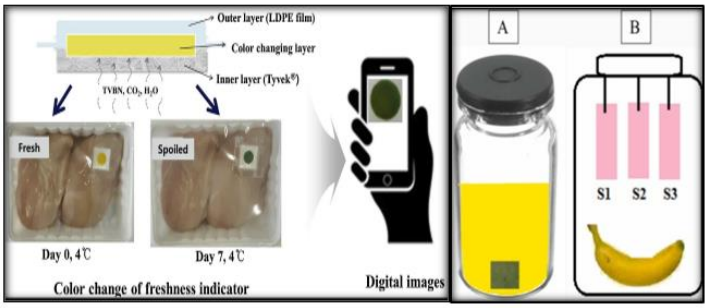

Fig 6. Triple layered freshness indicator, preventing migration from sensing element for food quality check

Usually, spoilage of seafood products such as fish, squid and shrimp are mainly caused by microbes. Microbial activity and enzyme degradation of proteins in seafood cause the release of unpleasant, off-flavour gases including ammonia, trimethylamine and dimethylamine, collectively represented as TVBN. The extent of the spoilage of the seafood product is directly reflected in the TVBN content. Accumulation of these volatile nitrogenous compounds in a headspace package elevates the pH and helps to know the change in the colour through naked eyes. No dye migration from sensing layer, which offers safe, simple and reliable, packaging solution with digital image processing via a smartphone for quality check**.**

Film properties

In the preparation of film. Film properties are very important which includes, mechanical properties (tensile strength etc), gas barrier properties, water vapour barrier, optical properties is through scanning electron microscope, aroma properties, thermal properties, water resistance properties. All these properties should meet to get good films. As it will be incorporated inside the food materials.

Conclusion

Past 40 years of transition in the food packaging, they have bought innovative technologies, such innovative technology is intelligent packaging. Till more studies using natural pigments are needed as the film will be in contact with the food where natural pigments are used instead of chemical substances as they are more hazards. Different technologies need to be developed to implement in large scale.

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