Ethylene Scavengers in Food Packaging Technology: A Review

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Abstract

Ethylene, being a plant hormone greatly effects the ripening, softening, color change and sensory properties of fruits and vegetable commodities. Use of ethylene scavengers as active packaging has been researched now from many years. Ethylene scavengers available in market mostly contain $KmNO_4$ as main functioning group embedded on various adsorbing materials. Ethylene scavengers are mainly classified on their mechanism of action: (1) catalysts, (2) ethylene reducing materials & (3) adsorbents. Further $KmNO_4$ being toxic materials is embedded on adsorbents. In past several years, some innovative ethylene scavenging materials have been researched. They include halloysite-nanotubes, natural clay particles, TiO_2, etc. Effects of ethylene scavengers include delaying of ripening, decrease in loss of chlorophyll content, delaying the pathogen and mold growth and also reduction in softening. Although, effects of ethylene scavengers depend on the type of ethylene scavengers used and the commodity of fruit and vegetables.

1. Introduction

It has been estimated that roughly one third of food produced for human consumption is lost or wasted globally, corresponding 50% of such food losses to F&V (Blanke 2014; FAO 2011). Blanke (2014) coined that, in the USA, food waste is reported to account for up to 30% compared to 16% in Europe, where 54% of the 82 kg versus 44 kg, composed of 26 kg vegetables and 18 kg fruit, of the reported food waste are fruit and vegetables with a relationship to the fruit ripening gas ethylene, while milk and meat products are exempt. On the other hand, Fruits & vegetable consumption has steadily augmented due to the increasing consumer profile interested in natural food products with high health-promoting properties. Nevertheless, the product selection by this consumer is primarily based on visual appearance attributes, such as good color, perfect shape and size, together with taste, aroma and texture (Hernández, *et al*., 2018).

To reduce the fruits and vegetable loss, storage management should take into account respiration rate, ethylene production and sensitivity of the product to several critical parameters such as storage temperature and determined gas concentrations (oxygen, carbon dioxide and ehtylene), while high relative humidity (RH) rates should be maintained with the aim of extending F&V shelf life (Kader 2005). In post-harvest phase, the quality risks are highly associated with factors such as commodity, cultivar, harvest type, handling, storage and supply (Singh *et al*. 2013).

Packaging is defined as enclosing food to protect it from tampering or contamination from physical, chemical and biological sources. Unlike traditional packaging, which must be totally inert, active packaging is designed to interact with the contents and/or the surrounding environment. (Prasad & Kocchar, 2014). Active packaging refers to the incorporation of additive agents into packaging materials with the purpose of maintaining or extending food product quality and shelf life (Lee *et al*., 2015). Active packaging as originally described by Labuza and Breene (1989) is used successfully to increase the shelf life of processed foods and meet consumer demands in terms of providing high-quality products that are also fresh and safe. At the present time, active and intelligent packaging systems are mainly used in Asia or the United States, whereas in Europe its use is not widespread (Prasad & Kocchar, 2014). One of the desired features of active food packaging materials designed for the storage of fruits and vegetables is the ethylene scavenging behavior (Tas, *et al*., 2017).

2. Ethylene

Ethylene gas is an odorless, colorless gas that exists in nature and is also created by man-made sources. (Gaikwad & Lee, 2017). Ethylene is a natural gaseous plant growth hormone which accelerates respiration of fruits and vegetables, induces fruit ripening, fruit softening and senescence even at low concentration (Abeles *et al*., 1992). It is synthesized mainly by a set of two enzymes called 1-aminocyclopropane-1-carbooxylic acid synthase (ACC synthase, ACS) and 1- aminocyclopropane-1-carbooxylic acid oxidase (ACC oxidase, ACO) (Singh *et al*. 2013; Lee *et al*., 2015). As fruits, vegetables and floral products mature, ethylene gas is released into their packaged environment (Gaikwad & Lee, 2017). Apart from the endogenous ethylene production by plant tissues, external sources of ethylene (e.g. engine exhausts, pollutants, plant, and fungi metabolism) occur along the food chain, in packages, storage chambers, during transportation, and in domestic refrigerators (Romero, *et al*., 1999)

In response to natural stresses and mechanical injury, the stress-responsive genes (Sharma *et al*. 2009; Yang *et al*. 2013) causes rapid and consistent cellular changes in perishable harvested produce (Singh *et al*. 2013). These changes are, possibly, mediated via 'stress ethylene' (Ansari *et al*., 2013). The level augmenting in response to numerous biotic and abiotic stresses and mechanical injury affects produce quality (Gapper *et al*. 2013), and therefore, ethylene synthesis occurs and action are of prime concern to manage produce at post-harvest phase.

Ethylene helps to accelerate ripening in fruit, followed by aging and ultimately death) It causes yellowing of vegetables, russet spotting on lettuce and has detrimental impact on shelf life of many fruits and vegetables (Zagory 1995). It also Causes Fruit ripening, accelerates fruit softening and aging, bacterial or fungal infection, mechanical or chemical damage and ultimately death (Lee *et al*.,2015 ; (Dobrucka, *et al*., 2017). Moreover, in both climacteric and non-climacteric fruit, ethylene can induce chilling injuries and physiological disorders (Wills, 2015). Ethylene is responsible for sugar content alteration, texture changes and volatile aromas synthesis. Meanwhile, in non climacteric fruit, ethylene stimulates senescence, often associated with yellowing of green tissues by promoting chlorophyll degradation and hastens to toughen and wilting (Barry and Giovannoni 2007; Lelievre *et al*. 1997; Saltveit 1999). Ethylene may increase pathogen susceptibility by inhibiting the formation of antifungal compounds, and in some cases, it can even stimulate the growth of fungi such Botrytis *cinerea Penicillium italicum* as on strawberries and on oranges (Abeles *et al*. 1992; Kader 2003).

3. Ethylene Scavengers

As for many other climacteric fruits, adequate control of ethylene concentrations inside the storage environment or suppressing the effects of ethylene is critical in order to extend storage life and maintain product quality. Ethylene scavengers are type of active packaging material that are used to scavenge ethylene from the packaging headspace of packaging. These ethylene capturing materials are usually used in the form of sachets that are placed inside the packaging, coating on packaging material or in form of ethylene scavenging active films (Tas, *et al*., 2017). Ethylene can be absorbed or adsorbed by a number of substances including activated charcoal, molecular sieves of crystalline aluminosilicates, Kieselguhr, bentonite, Fuller's earth, brick dust, silica gel (Kays and Beaudry, 1987) and aluminum oxide (Goodburn and Halligan, 1987).

Based on their mechanism of action, ethylene scavengers can be classified as,

(1) **Catalysts**: Often based on platinum/alumina, these operate at elevated temperature (> 200ºc) and catalytically oxidise ethylene to carbon dioxide (CO2) and water.

(2) **Stoichiometric oxidising agents**: Mostly based on potassium permanganate ($KMnO_4$), which again oxidizes ethylene and is itself reduced.

(3) **Sorbents**: These materials work by sorption of the ethylene and are often based on high surface area materials, including activated carbon, clays and zeolites. (Smith *et al*.,2009; Prasad & Kocchar, 2014)

Ethylene scavengers can reduce the ethylene generation-rate by blocking the binding site (Lee *et al*., 2015). 1-MCP has been reported to effectively delay the respiratory climacteric, the peak of ethylene production, mesocarp softening and skin colour change of avocado (Watkins, 2006) and it is used commercially in various avocado-producing countries. The ethylene binding inhibitor is believed to bind irreversibly to ethylene receptors at very low concentrations, blocking or delaying the maturation processes normally induced by ethylene (Meyer & Terry, 2010). As inhibitors of ethylene biosynthesis and action, good results have been found with polyamines and 1-methylcyclopropene (1-MCP) in terms of maintenance of fruit and vegetable quality and extension of postharvest shelf-life (Smith *et al* 2009;Prasad & Kocchar, 2014).

Many researchers have studied different types of ethylene scavenging systems and their effect on fresh commodities (Wills and Warton, 2004; Eastwell, *et al*., 1978; García *et al*., 2012; Abe and Watada, 1991; Martínez-Romero, *et al*., 2009). Although, the efficiency of those ethylene scavengers is significant they are toxic on some level. In that case a safe and efficient ethylene scavenging system is required to be developed. Such ethylene scavengers are described in the review paper which have been researched over past several years.

3.1 Stoichiometric oxidizing agents

Potassium permanganate widely used ethylene absorber that oxidizes ethylene to ethanol and acetate via the breakdown of ethylene's double bond (Lee *et al*. 2015). Though some CO_2 and water are produced, some partially oxidized species such as carboxylic acids may also be formed. As, potassium permanganate oxidizes ethylene its color changes from purple to brown, and thus, a color change indicates its residual ethylene absorbing capacity, but because of its toxicity potassium permanganate cannot be used in direct contact with food.

Chaves *et al*. (2007) showed that addition of KMnO4 in the MAP of sugar apple in PVC films delayed fruit maturity, increased the period for fruit to reach the maximum solid solutes content but, increased the pH and decreased the titratable acidity when it reached the consumption moment after 12 days of storage at 16°C. These discussions indicated that addition of Ethylene Scavengers (ES) in MAP controlled the ripening of fruit during storage and showed no negative effect on the fruit maturity when required for marketing or consumption.

Mortzavi & Karami (2015) conducted study to evaluate the effects of different packaging methods (control, passive MAP, MAP + ethylene scavenger sachet) and then storing at 5 or 15°C on the quality of khalal date fruit cv. barhee. To make the ethylene absorbing sachet, the super absorbent powder (sodium polyacrylate) was soaked in 5% KMnO4 solution, then 10 g of produced gel was cut and covered with a filter paper. Sachet was attached to the upper inside part of the package. MAP treatment with ethylene scavenger sachets was found to be effective in lowering weight loss (0.42%) and SBS (35%) and maintaining tissue firmness. Furthermore, other quality parameters experienced low changes in this treatment. Among the two storage temperatures, fruit which held at 5°C, remained firmer and showed superior in quality than those stored at 15°C.

Dobrucka *et al*., (2017) prepared an ethylene scavenger with the use of potassium permanganate. The prepared ethylene scavenger was used extend the storage time of fresh tomatoes. The ethylene scavenger was prepared by dipping activated zeolite in the saturated solution of potassium permanganate (6.4 g/100 mL) at 20°C for varying times, between 3 min and 6 h, allowing it to dry in air for 30 min, and repeating the dipping and drying process for up to 6 times. This test used three equal groups (five replicates per group) and placed them in commercial plastic packaging. The first group of tomatoes was placed in packaging without any ethylene absorber, while the other two groups were placed in packaging with any ethylene absorber. The sachets placed in the packaging contained respectively 1 and 2 grams of previously prepared ethylene absorber. The first mold on the tomatoes in the packaging without ethylene absorber occurred on the ninth day of the experiment. In the two remaining packets, mold appeared after nearly fifty days.

3.2 $KmNO_4$ supported on adsorbents

Ethylene removal media are usually composed of pellets of porous solids such as activated alumina, vermiculite, and silica gel that have been impregnated with potassium permanganate (KMnO4). The alumina functions primarily as the absorptive surface to trap the molecules of gas and is a carrier for the permanganate. Potassium permanganate is a broad-spectrum oxidizing agent that reacts with ethylene as well as with other contaminant gases. When it reacts by oxidizing ethylene to ethylene glycol, a visible color change occurs: fresh KMnO4 medium is purple in color but, after reacting with ethylene, it turns brown. The color change occurs first on the pellet's surface and eventually penetrates the core, indicating the reactive capacity is nearing exhaustion (Brody A.; Strupinsky E. and Kline, L., 2001).

Potassium permanganate due to their toxicity are generally embedded on different adsorbing materials such as porous inert material with a high surface area such as clays, silica (SiO_2) gel, zeolites, alumina (Al_2O_3), vermiculite and activated carbon (Hernández, *et al.*,2017). A number of oxidizers have been combined with adsorbents to remove the adsorbed ethylene such as potassium dichromate, potassium permanganate, iodine pentoxide, each respectively embedded on silica gel (Dobrucka, *et al.* 2017). Furthermore, some of the adsorbing materials can adsorb ethylene creating an adsorption–oxidation system, where the support material adsorbs ethylene and permanganate (MnO_4^-) oxidises it (Pathak *et al.* 2017).

Singh and Giri (2014) demonstrated that $KMnO_4$ embedded onto SiO_2 crystals could prolong shelf life of guava fruit (up to 7 weeks), under active packaging using low-density polyethylene (LDPE) film at 8 °C. With the use of $KMnO_4^-$ based ethylene scrubbers supported onto SiO2, minor changes in fruit firmness, total soluble solids content (SSC), titratable acidity (TA) and colour were obtained. In a recent study, it was reported that KMnO4 embedded onto

SiO2 crystals is a good tool to slow down the ripening and senescence process of 'Kajli' pointed gourd fruit (Bhattacharjee and Dhua 2017).

Murmu and Mishra (2018) In modified atmosphere packaging of guava, moisture scavenger (MS) sachet containing 30–50 g of coarse silica-gel and ethylene scavenger (ES) sachet containing 0–4 g of potassium permanganate was added as per central composite rotatable design. The coarse silica gel and potassium permanganate (98.5% purity) were procured from Merck (Mumbai, India) were used as MS and ES, respectively. The quadratic term of ES exhibited significant positive on the greenness value of guava stating that higher mass of ES kept guava greener and more unripe till 30 days of storage under MAP at low temperature. After two days of transferring guava at 30 °C following 30 days of actively packaged storage, the guava stored at 4 °C with 3 g ES and 46 g MS were least affected by chilling injury as they had the brighter color, lower firmness and the highest number of acceptable (95.33%) guava compared to other treatments.

Javanmard (2015), in his study, initially, produced ethylene scavengers by impregnating zeolite with potassium permanganate with zeolite clinoptiolite available in iran. After that, these ethylene scavengers were placed in the package of edible mushrooms and then transferred to the refrigerator with temperature 4℃ for a period of 20 days. The results showed that zeolite by concentration of 5%, 7.5% and 10% potassium permanganate and mesh 60 have a higher efficiency.

Spricigo *et al*., (2017) developed novel tools/devices to monitor and oxidize ethylene. Here, they proposed nanoscaled platforms based on silica (SiO_2) and alumina (Al_2O_3) nanoparticles impregnated with potassium permanganate ($KMnO_4$) that use color changes to indicate ethylene removal. SiO2 and Al2O3 in the microscale and nanoscale were impregnated with varied concentrations of KMnO4 through a simple mixture route, which systems were capable of oxidizing the ethylene in a closed atmosphere under relative humidity of 45, 60, 75, and 90%. The nanoscaled platforms impregnated with KMnO4 were capable of scavenging ethylene more efficiently for 1 h exposure. Additionally, the color changes experienced by the nanoscaled platforms, arising from the chemical reduction of potassium permanganate, function as an indicator of ethylene removal, which is particularly suitable for postharvest application.

In Korea, Orega ethylene scavengers were developed. Film made incorporating the Orega compound is claimed to have an ability to scavenge ethylene at a rate of at least 0.005 ppm per hour per square meter. The ethylene adsorptive activity of this film results from adding into the film a fine porous, inorganic material containing a large number of fine pores, 2 to 2,800 Å in size, such as pumice, zeolite, active carbon, cristobalite, and clinoptilolite. The finely divided porous material is sintered with a small amount of a metal oxide before being added to the film. The fine porous material is then incorporated into the plastic polymer for the film by conventional methods, and the polymer is extruded into the film. The particle size of the fine powder should be at least 200 mesh, and at least 1% by weight should be contained in the film. Treating the porous mineral with oxygen enhances the ethylene absorbing activity (Brody A.; Strupinsky E. and Kline, L., 2001).

The film containing the fine porous material not only has ethylene scavenging activity, but reportedly also has excellent permeability to gases such as oxygen, carbon dioxide, nitrogen, ethylene, and water vapor. Consequently, ethylene gas is discharged outside the film wrapping vegetables and fruit, and the inside of the film is maintained at a suitable relative humidity (Choi, 1991).

Another recent investigation by Ali *et al*. (2015), on the influence of different packaging materials and ethylene absorbent on biochemical composition, antioxidant and enzyme activity of apricot, made it quite evident that apricot harvested at commercial maturity stage and packed with low density polyethylene films along with ethylene scavenger (KMnO4) can be successfully stored at ambient conditions up to two weeks.

A Hungarian paper manufacturer, Dunapack, has developed paper called "Frisspack" for manufacture into corrugated fibreboard cases to prolong the shelf life of contained fresh produce. A component of their "Frisspack" package tries to bond with the ethylene generated during fruit and vegetable ripening in order to decrease the rate of respiration. To bond with ethylene, the most commonly used materials are silica gel, with potassium permanganate compounds to react with the ethylene. According to Dunapack, silica gel adsorbs ethylene while KMnO4 oxidizes it (Brody A.; Strupinsky E. and Kline, L., 2001).

Frisspack paper was developed to chemically bond with ethylene released during fresh and minimally processed produce respiration. Dunapack achieved this objective by dispersing a chemosorbent of small particle size (average particle size 1 μm) with high absorption capacities among the fibers in the early phase of the paper production, when the fibers are still in approximately 1% water suspension. The chemosorbent can be uniformly blended among the fibers and have surface characteristic features such that the particles connect to the paper fibers by hydrogen bonding. By this means, the chemosorbent particles are embedded in the paper so firmly that they do not become separated from the paper due to mechanical action or chemical effects (e.g., juice leakage from fruits during storage) (Brody A.; Strupinsky E. and Kline, L., 2001).

Szikla (1993), has studied the effect of Frisspack package materials on the ripening of fruits was tested with a wide variety of fresh produce products, including apples, pears, peaches, apricots, bananas, cherries, grapes, strawberries, raspberries, carrots, onions, potatoes, mushrooms, tomatoes, and green peppers. Frisspack paper significantly retarded the ripening of almost all types of fruits and vegetables tested. The loss in weight and sugar content of the fruits stored in packages made of Frisspack paper was less, and they retained their texture and color to a greater extent than the fruits stored in conventional packaging materials.

Best D. *et al.* (2005) has studied the storage of the ‘Nijisseiki’ cultivar of Japanese pears over three seasons for periods up to 36 weeks at 0°C. Storage in 50 pm thick low-density polyethylene (LDPE) bags at 0°C considerably delayed yellowing in all experiments, even after fruit was removed to 20°C for 1 week at the end of storage. The addition of an ethylene absorbent made from potassium permanganate on aluminium oxide (Purafil II) further delayed yellowing. Carbon dioxide levels in both treatments varied, but were generally in the range 2-3%. Oxygen levels remained high, generally 16-19%. In bags without Purafil, ethylene levels rose slightly during storage and were generally about 0.15 pi I 1. When Purafil was included in the bags, the ethylene level was reduced 10-fold or more. A sensory test indicated that the use of LDPE bags and ethylene absorbent resulted in fruit with better eating quality than fruit stored in air. Disorders over the 3-year investigation were low even after long-term storage. The use of polyethylene bags reduced the severity of flesh browning, and flesh spot decay was virtually absent. Tire use of bags increased the severity of core browning. Inclusion of an ethylene absorbent in bags reduced the severity of disorders, particularly core browning. Treatment of the fruit with 1 - methylcyclopropene (1-MCP), before or during storage, resulted in higher ethylene levels in the polyethylene bags. At the concentrations used, 1-MCP did not improve the storage of ‘Nijisseiki’ compared to the use of polyethylene bags with Purafil II.

3.4 Activated Carbon Ethylene Removers

Abe and Watada (1991), has studied the effect of metal catalysts on activated carbon remove ethylene from air passing over the bed of activated carbon. Activated charcoal impregnated with a palladium catalyst and placed in paper sachets effectively removes ethylene by oxidation from packages of minimally processed kiwi, banana, broccoli, and spinach.

Japan's Honshu Paper offers the "Hatofresh Systems," which are based on activated carbon impregnated with bromine-type inorganic chemicals. The carbon-bromine substance is embedded within a paper bag or corrugated fibre-board case used to hold fresh produce. They claim that the bag will scavenge 20 cc of ethylene per gram of adsorbent by the combination of ethylene with bromine (Brody A.; Strupinsky E. and Kline, L., 2001).

Japan's Mitsubishi Chemical Company produces "SendoMate," which employs a palladium catalyst on activated carbon that adsorbs ethylene and then catalytically breaks it down. The product comes in woven sachets that can be placed in packages of produce (Brody A.; Strupinsky E. and Kline, L., 2001).

Activated carbon-based scavengers with various metal catalysts can also effectively remove ethylene. They have been used to scavenge ethylene from produce warehouses or incorporated into sachets for inclusion into produce packs or embedded into paper bags or corrugated board boxes for produce storage. A dual-action ethylene scavenger and moisture absorber has been marketed in Japan by Sekisui Jushi Limited. Neupalon™ sachets contain activated carbon, a metal catalyst and silica gel and are capable of scavenging ethylene as well as acting as a moisture absorber (Abeles *et al.*, 1992; Rooney, 1995).

3.5 Activated Earth Ethylene Removers

Japan's Nissho suggests a film that incorporates finely ground coral (primarily calcium carbonate), having pore sizes in the range of 10 to 50 μm. After incorporation into a polyethylene film, the coral is claimed to absorb ethylene (Brody A.; Strupinsky E. and Kline, L., 2001).

"BO Film," marketed by Japan's Odja Shoji, is a low-density polyethylene film extruded with finely divided crysburite ceramic, claimed to confer ethylene-adsorbing capacity (Brody A.; Strupinsky E. and Kline, L., 2001).

Zagory, in his section on ethylene removal in the book *Active Food Packaging* (Rooney, 1995), asserts that the evidence offered in support of claims about ethylene removal by activated earth or clay is generally based on shelf life experiments comparing common polyethylene film bags with clay-filled film pouches. Such evidence generally shows an extension of shelf life and/or a reduction of headspace ethylene. Although the finely divided clays may adsorb ethylene gas, they can also create pores within the plastic bag and alter the gas-transmission properties of the bag. Because ethylene diffuses more rapidly through open pore spaces within the plastic than through the plastic itself, ethylene would be expected to diffuse out of these pouches faster than through pristine polyethylene film bags. In addition, carbon dioxide within these pouches is transmitted more rapidly and oxygen enters more rapidly than with a comparable conventional polyethylene film pouch due to the gaps in the film. These effects can enhance shelf life and reduce headspace ethylene concentrations independently of any ethylene adsorption. In fact, almost any powdered mineral can confer such effects without relying on expensive Oya Stone or other specialty minerals.

4. Ethylene Adsorbers

Ethylene scavengers which adsorb ethylene for to remove it from packaging materials are generally porous structures which include activated alumina, activated charcoal and different types of clays. Such ethylene binding clays include, silica, diatomaceous earth, anti-caking agent (bentonite), China clay, quartz, Oya stone in powder form, natrolite, ozone (Rodríguez, *et al.* 2014). They possess higher surface area and are of micro &/or nano structure.

The role played by the surface area size, the success of a scrubber also depends on the material type and other physical characteristics such as shape and ethylene adsorption ability. For example, it has been reported that $KMnO_4$ based ethylene scrubbers supported onto Al_2O_3 nanoparticles have higher ethylene removal rate than scrubbers based on SiO_2 nanoparticles (Spricigo *et al*. 2017). In addition, many other parameters also play a key role in the performance of a scrubber product, e.g. temperature and RH (Gaikwad and Lee 2017; Keller *et al*. 2013).

Rodríguez *et al*. (2014) studied the efficiency of a Chilean natural zeolite (NZ-Ch) against a commercial Na+ montmorillonite (Cloisite Na+). The rate constant of the ethylene adsorption was nearly double for NZ-Ch compared with MtNa+. Maximum adsorption capacity reached values of 5.4 µl g-1 for NZ-Ch and 1.28 µl g-1 for MtNa+. Films of low-density polyethylene (LDPE) were obtained with different NZ-Ch concentrations. After 50 hours, a removal of 37% of ethylene present on headspace was achieves with 10% of ZN-Ch in LDPE active films.

5. Nano-halloysite tubes

Recently, naturally found porous materials like montmorillonite, cloisite, zeolite, clay, and Japanese oya have gained much attention as ethylene scavengers for packaging applications. Halloysite nanotubes are used as a substitute for traditional ethylene scavengers in applications where cost is concern HNTs are natural aluminum silicate nanoparticles presenting hollow tubular nanostructures (Tas, *et al*., 2017). Halloysite nanotubes are “green” materials mined from natural deposits (Bodbodak and Rafiee 2016), and they are not harmful to the environment and are generally recognized as safe for food packaging by the US Food and Drug Administration (Lee *et al*. 2017). Their low price and high performance facilitate the industrialization of halloysite nanotubes—polymer nanocomposites. Gaikwad *et al,* (2018). Halloysite nanotubes have high aspect ratio that allows them to be used as nanocarriers and can be efficiently dispersed in the polymer matrix, resulting in the release of the active agent in nanocomposite (Yuan *et al*. 2015).

Gaikwad *et al,* (2018) tested the effect of storage conditions on the kinetics of ethylene adsorption. Raw halloysite nanotubes were subjected to alkaline treatment to increase their pore size. They indicate that NaOH increased the pore size in the treated halloysite nanotubes (Fig.1). They compared the efficacy of raw halloysite nanotubes versus alkaline halloysite nanotubes. Results show that alkali-treated halloysite nanotubes have the highest ethylene adsorption capacity at 11% relative humidity and 23 °C. The cell containing alkaline halloysite nanotube sample showed speedy adsorption of ethylene, the rate constant of the ethylene adsorption was 0.7107 min^{-1}. After 24 h, 49 µL of ethylene gas present in headspace was removed with 1 g of alkali halloysite nanotubes. It can be concluded that the high ethylene adsorption capacity of alkaline halloysite nanotubes was associated with their improved pore structure, which enhanced adsorption of ethylene.

Tas, *et al*. (2017) utilized HNTs as nanofillers absorbing the naturally produced ethylene gas that causes softening and aging of fruits and vegetables; at the same time, limiting the migration of spoilage-inducing gas molecules within the polymer matrix. HNT/polyethylene (HNT/PE) nanocomposite films demonstrated larger ethylene scavenging capacity and lower oxygen and water vapor transmission rates than neat PE films. Nanocomposite films were shown to slow down the ripening process of bananas and retain the firmness of tomatoes due to their ethylene scavenging properties. Even used at 1, 3, and 5 wt.% HNT did not have significant variations in mechanical properties compared with neat PE films. 5 wt.% HNTs had a 0.067% ethylene adsorption capacity at 1 bar pressure. 1g of food packaging film had 0.56 ml ethylene adsorption capacity.

6. Ethylene scavenging by catalysts

Often based on palladium these operate at elevated temperature (> 200ºC) and catalytically oxidise ethylene to carbon dioxide (CO_2) and water. At the time of their function they are not utilized, thus they give a long-term performance with significant reduce of ethylene from the surrounding environment.

Smith *et al*. (2009) described a novel palladium-promoted zeolite material with a significant ethylene adsorption capacity at room temperature. Ethylene adsorption capacity measurements were carried out at room temperature (21ºC) in a plug flow reactor using 0.1 g of active Pd-based material with a gas composition of 200 μl l^{-1} ethylene, 10% (v/v) oxygen balanced with helium, at a flow rate of 50 ml min^{-1}, with and without ca. 100% relative humidity (RH). The Pd-based material typically removed all measurable ethylene until breakthrough occurred. The results are similar with the results obtained by Debrucka, *et al*., (2017).

Meyer & Terry (2010) in their study reported on the effect of 1-methylcyclopropene (1-MCP) and a newly developed palladium (Pd)- promoted ethylene scavenger (e + ®Ethylene Remover) on changes in firmness, colour, fatty acids and sugar content of early and late season avocado (*Persea americana Mill*.), cv. Hass, during storage at 5 C and subsequent ripening at 20 C. The e + ®Ethylene Remover effectively delayed ripening of avocado stored at 5 C. 1-MCP was more effective at inhibiting ripening, but, in contrast to e + ®Ethylene Remover, it impaired subsequent ripening.

Activated charcoal impregnated with palladium catalyst is also used to scavenge ethylene from fresh produce (Biji, *et al*., 2015). Recently, a newly developed palladium Pd-promoted material was shown to be effective at removing ethylene at low temperatures to sub physiologically active levels and, accordingly, to effectively delay the climacteric-induced ripening of avocado cv. Hass fruit (Terry *et al*., 2007). The material consisted of a zeolite impregnated with finely dispersed Pd particles (Smith, *et al*., 2009).

Bailén *et al*. (2007) studied the ethylene adsorption capacity of different types and masses of activated carbon, to predict the performance of the ethylene adsorption process, to improve the removal of ethylene by impregnating granular activated carbon (GAC) with palladium, and to analyse the effect of this product on the removal of ethylene released from tomatoes. In an *in vitro* system, both GAC and powdered activated carbon (PAC) effectively absorbed exogenous ethylene; GAC was the most effective. The percentage ethylene removal achieved by GAC after 48 h was 70.10 ± 1.05%, but this increased to 84.69 ± 0.79% with GAC-Pd.

In order to remove ethylene surrounding the broccoli during shelf-life Cao, *et al*. (2014) developed a novel ethylene scavenger supported by acidified activated carbon powder (AACP). Results showed that acidification of the supporter might have a profound effect on ethylene scavenging. Ethylene removal rate of the palladium chloride ($PdCl_2$)-impregnated AACP scavenger dramatically increased and was further promoted with the addition of copper sulfate (CuSO4). The novel scavenger was optimized and prepared with AACP containing 10 mg/g PdCl2, 30 mg/g CuSO4 and less than 300 mg/g moisture. Its maximum ethylene removal capacity was 21.77 mL/g at 25˚Cor 20.18 mL/g at 5◦C. The ethylene removal capacity could be regenerated by 81.6% by heating at 175˚C for 20 min. The ACCP–$PdCl_2$–$CuSO_4$ ethylene scavenger could significantly delay yellowing and quality loss of broccoli and thereby extend the shelf-life at 20◦C for 2 d. Ethylene concentration in the stored environments was decreased to 0.21 µL/L or less from 0.91 µL/L during shelf-life by the scavenger.

Titanium dioxide (TiO2) is a chemically inert material and is widely used in various food, medical and biological products. TiO2 has been reported to exhibit antimicrobial and ethylene photo-degradation activity when exposed to UV light by generating hydroxyl radicals (OH•) and reactive oxygen species (ROS) on its surface which further reacts with organic molecules. There are also reports of the use of photocatalytic oxidation of ethylene using titanium dioxide (TiO2), which can occur at room temperature (Smith *et al*., 2009). Also, TiO2 interactions with ripening gas (ethylene), slow down the maturation rate of climacteric fruits and extend the shelf life of ready to use fruit and vegetables (Costa *et al*., 2011). TiO2 has the unlimited ethylene scavenging capacity, because TiO2 is not being consumed in the reaction (Shankar & Rhim, 2014).

It has been approved by the American Food and Drug Administration (FDA) for use in human food, drugs and cosmetics and compounded in food contact materials such as cutting board, plastic packaging and other surfaces in contact with unprotected food (maximum allowable amount for food is 1% TiO2 (Ghanbarzadeh, *et al*., 2014).

Karbach *et al*. (2015) A phenoxy-imine titanium catalyst (FI-catalyst) for selective ethylene trimerization was immobilized on MAO-pretreated silica and its activity and selectivity was compared with that of the corresponding homogeneous catalyst system. The homogeneous and heterogeneous ethylene oligomerization was conducted in the presence of different aluminum alkyls, commonly used as scavengers during olefin polymerization to remove residual oxygen and moisture from the reaction medium. Both the homogeneous and heterogeneous catalysts were strongly affected by the presence of scavenger in the reaction medium. Upon activation with R_3Al/MAO (R= Et, *n*Oct, *i*Bu), the homogeneous catalyst switches selectivity from ethylene trimerization to polymerization. NMR spectroscopic investigations indicate that this change of selectivity can be attributed to ligand exchange between the pre-catalyst and the aluminum alkyl. The thereby formed ligand-free titanium alkyls act as polymerization catalysts and are responsible for the increasing polymer formation. Using the heterogeneous catalyst, the scavenger employed during ethylene trimerization was found to be of crucial influence regarding the activity of the catalyst and the occurrence of reactor fouling.

Kaewklin *et al*., (2018) further studied the feasibility of active packaging from chitosan (CS) and chitosan containing nanosized titanium dioxide (CT) to maintain quality and extend storage life of climacteric fruit. The CT nanocomposite film and CS film were fabricated using a solution casting method and used as active packaging to delay ripening process of cherry tomatoes. Tomatoes packaged in the CT film evolved lower quality changes than those in the CS film and control. The results suggested that the CT film exhibited ethylene photodegradation activity when exposed to UV light and consequently delayed the ripening process and changes in quality of the tomatoes.

7. Effect of Ethylene Scavenger Packaging on Shelf-life and quality of Fruits and Vegetables

Use of ethylene scavengers to remove ethylene from the packaging environment also effects quality of fruits and vegetables. The reduction in ripening and deterioration, extension of shelf life has been reported (Lee *et al*., 2015). Bailen *et al.* (2006) reported a reducing the rate of softening in tomatoes. The use of GAC-Pd to remove ethylene from the environment could help to delay ripening and extend the time that tomatoes are acceptable for consumption (Bailén *et al*. 2017). Results from the experiments on edible mushrooms proved that impregnated zeolite with potassium permanganate have a meaningful influence in prevent the weight loss, decrease of moisture content and L-value, increase of a-value and overall color change (ΔE) and decrease of firmness texture of mushrooms (Javanmard, 2015).

Ethylene gas caused chlorophyll degradation and de-greening of the guava peel color. The use of ethylene Scavenger arrested ripening and prevent the chlorophyll loss. (Murmu & Mishra, 2018). An ethylene scavenging system, containing active carbon and palladium (II) chloride as a catalyst, effectively reduced the rate of softening of minimally processed kiwi fruits and bananas and decreased the loss of chlorophyll in spinach leaves stored at 20°C by absorbing ethylene from the medium (Abe and Watada 1991).

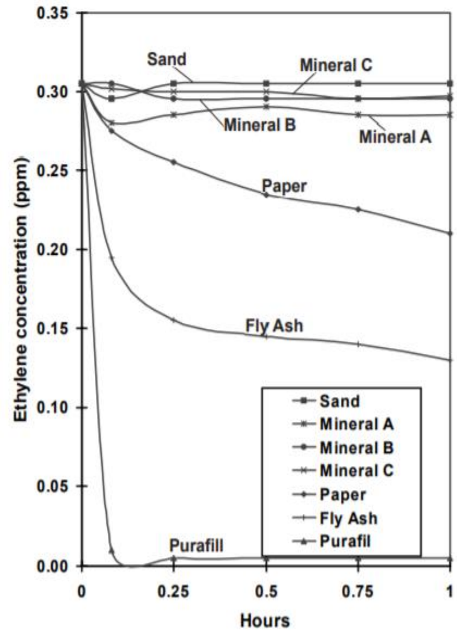
Fruit packed together with kg $^{-1}$ Ethylene scrubbers of $kmno_4$-sio_2 (8 g of fruit) in PP bags at 29–33 °C showed lesser changes in sensory properties and lowered chlorophyll content decrease compared to fruit without Ethylene scrubbers. It also delayed the growth of mold on tomatoes (Dobrucka, *et al*., 2017).

It may not be possible that effect of each type of ethylene scavenger on the retention of quality. Mansourbahmani *et al*., (2017) studied efficiency of ethylene scavengers on the maintenance of postharvest quality of tomato fruit. The objectives of the study were to achieve the best level of each ethylene scavenger and evaluate the effect of selected levels of treatments on some quality traits of tomato during storage. Tomato fruits were subjected to four levels of treatments: palladium-promoted nano zeolite, KMnO4- promoted nano zeolite, 1-MCP, CaCl2, salicylic acid (SA) and UV-C. The sampling was done at 0, 7th, 14th, 21st, 28th and 35th days of cold storage. The results showed that palladium-promoted nano zeolite 5%, KMnO4- promoted nano zeolite 20%, 1-MCP 30 ppm, CaCl2 2%, SA 1% and UV-C 15 min levels had the most ethylene scavenging function. Effectiveness of the treatments in ethylene scavenging was in the order: palladium > KMnO4 > 1-MCP > SA = CaCl2 > UV-C. The palladium promoted nano zeolite 5% had more positive effects on phenol content, polygalacturonase activity, lycopene content, fruit firmness and weight loss, and UV-C 15 min had effect on decay severity as compared to the other treatments. Overall, palladium-promoted nano zeolite 5% could be considered not only as favorable tool in tomato shelf life extension but also in preservation of quality characteristics of tomato fruits during storage. Moreover, the UV-C 15 min treatment could be an effective method for reducing decay severity and maintaining postharvest quality of tomato fruits.

8. Novel approaches to ethylene-removing packaging

The most promising new development in ethylene-removing packaging is the use of electron-deficient dienes or trienes incorporated in ethylene-permeable packaging. The preferred diene or triene is a tetrazine. However, since tetrazine is unstable in the presence of water, it must be embedded in a hydrophobic, ethylene-permeable plastic film that does not contain hydroxyl groups (Holland, 1992). Appropriate films would include silicone polycarbonates, polystyrenes, polyethylenes and polypropylenes. Approximately 0.01-1.0 M dicarboxyoctyl ester of tetrazine incorporated in such a film was able to effect a ten-fold reduction in ethylene in sealed jars within 24 h and a 100-fold reduction within 48 h (Holland, 1992). The tetrazine film has a characteristic pink color when it is new and turns brown when it becomes saturated with ethylene so it is possible to know when it needs replacing (Holland, 1992).

Reid, M. and Dodge, L; (1995), has studied and compare the effects of different ethylene scavengers. They have tested range of materials including three similar mineral materials, an activated carbon impregnated paper, and a reprocessed fly ash material. As controls, they used sand, which is effectively inert, and Purafil, a purple granular material containing potassium permanganate which is known to be a very effective ethylene absorbent. They placed samples of the different materials in small sealed jars, then injected enough ethylene to give a concentration of about 0.3 ppm. the Purafil absorbed the ethylene in the jar almost immediately, reducing the ethylene concentration to near zero. the three mineral materials absorbed no ethylene at all and were no different from sand. Ethylene levels in the jars containing any of these four materials remained near 0.3 ppm for the hour shown in the graph and, when tested 24 hours later, were still near 0.3 ppm. The paper material and the fly ash absorbed ethylene slowly, resulting in a reduction in ethylene concentration in the jars of one-third and one-half, respectively, over the one-hour test period.



9. Conclusions

Ethylene is the main ripening inducing agent of fruits and vegetables and may cause the premature ripening of some products, even ruin others. In order to reduce the concentration of ethylene and extend the shelf life of horticultural products, solutions already existing in the market, whose only use limitation is related to the material to be used, whether it is approved for food contact have been developed. If we want to use a scavenging compound for direct food contact, we must seek suitable solutions for this contact that are mainly limited to solutions inhibiting and reducing the ethylene concentration.

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