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An iodine supplementation of tomato fruits coated with an edible film of the iodide-doped chitosan



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ABSTRACT

In general, the risk of numerous thyroid cancers inevitably increases among people with iodine deficiencies. An iodide-doped chitosan (CT-I) solution was prepared for dipping tomatoes to coat the fresh surface with an edible film (1.5 μ m), thereby providing iodine-rich fruits for daily intake. Characterisation of the thin film was conducted by FTIR and SEM. Stability of the CT-I film was studied via water immersion at various time intervals, and no residual iodide leached out due to intrinsic interactions between the cationic amino group of chitosan and iodide ions. Moreover, the iodide supplement exhibited no effect on the antioxidant activity of tomatoes. The iodine content in the film-coated tomato was determined by ICP-OES. The tomato coating with 1.5% (w/v) CT-I contained approximately 0.4 μ g iodide per gram fresh weight. In addition, the freshness and storability of iodine-doped tomatoes were also maintained for shelf-life concerns.

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1. Introduction

lodine is a vitally important nutrient as a trace element detected in both organs and tissues for biological and physiological functions (Rana & Raghuvanshi, 2013; Leufroy et al., 2015). lodine is essential for an efficient metabolism and thyroid functioning, and thyroid hormones are involved in growth, development and the control of certain metabolic processes in the body (Anonymous, 2002; da Silva et al., 2016). Although iodine deficiencies result in iodine deficiency disorders (IDDs), the most well-known disorder is goiter, which is an enlargement of the thyroid gland. The various IDDs affect approximately 35% of the world population (WHO, 2004, 48pp). Worldwide, it is estimated that over

two billion people have insufficient iodine intake and are at risk for iodine deficiency (Zimmermann, 2009; Shelor & Dasgupta, 2011; Nitschke & Stengel, 2015). In Thailand, health agencies generally state that most people are iodine deficient because low iodine status is related to numerous diseases, including thyroid cancer. Thyroid cancer is among the top ten leading cancers in females with an age-standardised incidence rate (ASR) of 1.0 in males and 5.4 in females (Vatanasapt, Sriamporn, & Vatanasapt, 2002). To avoid iodine deficiency and its associated disorders, its concentrations thus need to be controlled on a daily uptake of approximately 180–200 µg. In fact, the thyroid gland may store iodine in its iodide form. Next, iodide could be reduced to iodine by thyroglobulin because the iodine cannot be utilised directly by the body. Therefore, iodide levels are generally necessary to be optimised in daily diets.

However, the use of iodine is limited by its volatility, which can be controlled by attaching it to polymers (Siggia, 1957; Moulay,



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2013). Because iodine is a powerful antimicrobial agent (Xing, Deng, & Yang, 2005; Shirai et al., 2011; Tang, Xie, Sai, Xu, & Ding, 2015), polymer-iodine complexes have gathered significant interest. These complexes have an interesting potential for use with fresh foods. Chitosan [β -(1-4)-2-amino-2-deoxy-D-glycopyranose] is a polysaccharide obtained from the alkaline deacetylation of natural chitin (Rabea, Badawy, Stevens, Smagghe, & Steurbaut, 2003; Kadokawa, Shimohigoshi, Yamashita, & Yamamoto, 2015; Mahé, Brière, & Dez, 2015), and its derivatives have been widely applied because of its several advantages (Morgado et al., 2013) due to its prominent active amino groups. Chitosan can also be functionalised to introduce positively charged N atoms and protonated amino groups for use in a variety of foods and food products (Kumar, Muzzarelli, Muzzarelli, Sashiwa, & Domb, 2004; Muzzarelli & Muzzarelli, 2005). Chitosan is edible, biocompatible, non-toxic, and non-polluting, and it offers good aesthetics in its appearance, good vapour barrier properties (Leceta, Peñalba, Arana, Guerrero, & De La Caba, 2015; Tang et al., 2015) and a low production cost. Edible chitosan coatings have traditionally been used to improve food appearance and maintain quality because they are considered eco-friendly (Khwaldia, Ferez, Banon, Desobry, & Hardy, 2004). Additionally, the market demand of ready-to-eat fruits and vegetables has witnessed a rapid expansion. What consumers perceive as the most appealing attributes of these products include their fresh-like appearance, and nutritiousness in addition to convenience.

In order to increase the efficient application of the edible chitosan film, likewise, it increases in the nutritional values of the tomato fruits by supplementing with iodide to avoid iodine deficiency. The objective of the current study was to develop chitosan-iodide (CT-I) coating as an edible film for fresh tomato samples, which offers the advantages of a nutritional supplement, easy availability and low cost materials.

2. Materials and methods

2.1. Chemicals and reagents

Shrimp shell chitosan (degree of deacetylation $\ge 75\%$) used for edible film preparation was purchased from Sigma–Aldrich (Japan). Glacial acetic acid was obtained from AnalaR (England). Potassium iodide (KI) and sodium carbonate (Na₂CO₃) were purchased from Carlo Erba (Italy). 2,2-Diphenyl-1-picrylhydrazyl (DPPH) and catechin were obtained from Sigma–Aldrich (U.S.A.). Folin–Ciocalteu reagent was acquired from Merck (U.S.A.). Ethanol was obtained from QRecTM (New Zealand). Chemicals and solvents used in this study were all analytical grade, and all aqueous solution were prepared with deionised water (Milli Q Millipore 18.2 M Ω cm⁻¹ of resistivity) by a Simplicity Water purification system, Model Simplicity 185, Millipore Corporation (U.S.A.).

2.2. Instruments

An iodide analysis was carried out by an Agilent Technologies Cary 60 UV–Visible spectrophotometer (Germany). A quartz cuvette with a light path length of 10 mm and a volume of 750 μ L was used for microscale measurements.

The iodide content in tomato samples was determined by a Perkin-Elmer OPTIMA 2100 DV inductively coupled plasma-optical emission spectrometer (ICP-OES) (Wellesley, Massachusetts, U.S.A.), with a standard ICP torch and peristaltic pump. The operational system is controlled with PE Winlab software. The instrument and operating conditions for ICP-OES are described in the Supplementary data, Table S1. Fourier transform infrared (FTIR) spectra of the CT-I film were recorded using an IR spectrometer (Tensor 27, Bruker Optics, Germany) and using an attenuated total reflection (ATR)-FTIR spectrophotometer. The transmission infrared spectra of all samples exhibited broad peaks in a range of $600-4000 \text{ cm}^{-1}$ at a resolution of 4 cm⁻¹.

Scanning electronic microscopy (SEM) images were obtained using a Hitachi S-3000 N scanning electron microscope. A PC was used to control the variable-pressure SEM with the ability to switch between the high vacuum and variable pressure modes. This microscope has a high density frame memory of 1280×960 pixels. The obtained thin film pieces were mounted on an aluminium grid, and the grids were later enabled to dry at room temperature, coated with gold and observed using an accelerating voltage of 10 kV.

2.3. Preparation of an edible film

Edible chitosan (CT) coating film was prepared by slowly dissolving chitosan in 50 mL of 1% (v/v) acetic acid solution and homogenised by constant stirring (Haramony, HTS-1003, Japan). The mixtures were treated overnight to ensure air bubbles were removed, which could affect thin film properties. Subsequently, the solution was filtered to remove the insoluble residue and poured into a Petri dish and dried in ambient conditions until a thin film formed. The thin film was peeled off the Petri dish for analysis. The thickness of the film was measured from SEM images.

The CT thin film was prepared with iodide as follows: chitosan powder was added into 50 mL of 1% (v/v) acetic acid under vigorous stirring. A 100 μ g mL⁻¹ potassium iodide solution was added slowly under continuous stirring at approximately 400 rpm for 4 h. Next, the resultant solution was poured into a Petri dish and left to dry for approximately 3–5 h in ambient conditions to create the CT-I film.

2.4. Dip-coating of the edible film on tomato fruits

Dip-coating real samples into the edible film solution was performed on mature cherry tomatoes (*Solanum lycopersicum*), which were selected based on the size (8.53 ± 2.19 g per fruit) and highquality colour, and the fruits were free from an injury or diseases. Before sample treatment, tomatoes were washed and allowed to air dry at room temperature. Next, the edible film coating was formed by a dipping method. The fruit samples were immersed in the iodide-doped chitosan solution for 1 min, and the excess was allowed to drain off. Next, the samples were placed on trays and allowed to dry at ambient temperature.

The four groups edible films were coated from solutions of (a) 0.5% CT, (b) 0.5% CT-I, (c) 1.5% CT and (d) 1.5% CT-I. An uncoated group of tomatoes was set aside as a control sample. Each of the coatings was performed in triplicate. Data were recorded for 7 days. Storage experiments were performed at ambient temperature. For each experiment, all tomato samples were purchased at the same time. The treatments and characterisations were also conducted simultaneously. The obtained results were compared within each experiment.

2.5. Ultrasonic-assisted extraction

The CT-I solution (approximately 0.1 mL) or the CT-I film (approximately 0.0025 g) was weighed accurately and extracted with deionized water by an ultrasonic-assisted extraction (UAE) for 5 min under a constant frequency of 35 kHz. The iodine extract in the supernatant was immediately determined spectrophotometrically after centrifuging at 5000 rpm for several min. The absorbance was measured at 225 nm (Ito et al., 2012).

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