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Detection of adulterated honey by surface plasmon resonance optical sensor

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ABSTRACT

In this study, surface plasmon resonance based on Kretschmann configuration is employed as an alternative method to detect adulteration of pure honey. The adulterated honey was prepared by diluting three types of sugar adulterants (fructose, glucose and sucrose) into pure honey. The concentration of each adulterant is tested from 2% until 10% adulteration with 0% adulteration represents the reference for pure honey. All the resonance angles of adulterated honey demonstrated similar behavior by shifting to smaller angle than pure honey. The measured sensitivities are 0.1266°/%, 0.1065°/%, and 0.0988°/% for fructose, glucose and sucrose adulterants, respectively. The shift of resonance angle as a function of adulterants concentration in pure honey was plotted with linear regression greater than 0.95 for all samples. The outcome has disclosed a real-time, rapid and non-destructive sensor to be promoted as well-developed honey sensor.

1. Introduction

Honey is a complex natural food with a lot of benefits to human. It made up of various chemical components, the main constitutes are carbohydrates including glucose, sucrose, fructose and maltose which constitute approximately 77% of the average honey [1]. The biochemical variation in the composition of pure honey is supposed to guarantee the originality taste and benefits. It is strictly said that the content of fructose and glucose together in honey should be not less than 60 g/100 g (60% in mass ratio) and sucrose content should be not more than 5% on mass basis even though amount of each composition is varied with several nature factors. Moreover, the Codex Standard for honey also states that honey sold as such shall not have added to it any food ingredient, including food additives, nor shall any other addition be made other than honey [2]. Therefore, any biophysical changes directly from the addition of foreign substances will forge the honey as a non-authentic food that leads to the taste quality issue. This situation usually happened because of the limited stock availability and high demand market that triggers the pure honey towards adulteration [3]. The issue of product integrity among the honey supplier community should be highlighted. The kind of this valuable food has to inherent quality of containing all

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the composition parts that meets the agreed specifications [4]. Thus, a rapid venture to explore and develop a device with specialty to detect honey fraud is significantly useful to prevent persistent problems on this issue.

When it comes on sample adulteration detection, optical method based on fiber optic is commonly utilized. From previous reports, this technique has been employed to determine; adulteration in a liquid sample [5], fuel adulteration [6], and adulteration traces in coconut oil [7]. An alternative method for honey adulteration detection, optical phenomenon of surface plasmon resonance (SPR) is proposed as it particularly pleading in food safety area since they may perceive analytes in complex matrices [8]. Moreover, this technique has sparked significant interests in food analysis lately [9,10]. SPR detection was able to perform on a mixture of several toxin serotypes in 20% honey solution [11] and also the chloramphenicol and chloramphenicol glucuronide residues in honey [12]. Thus, SPR is expected to be able to detect any foreign substances addition in pure honey. The SPR signal is induced from the interaction between incident light and a metal layer by generating surface waves that propagate along the interface between these two media. Normally, the metal layer is placed on a piece of prism based on the Kretschmann model [13]. The angle of incident light is controlled through a rotation stage and the reflected light from the prism is detected. At a specific angle, when the surface wave is generated, the reflected light is significantly attenuated owing to the energy transfer mechanism [14]. This measurement can be determined from the signal dip that corresponds to the refractive index of target material on the metal layer. This SPR signal is very sensitive to the change of refractive index on the metal layer [15].

Since SPR is a non-destructive method, it is suitable for various applications in biology and chemistry fields [16]. Therefore, the material composition is not influenced by this technique and it becomes one of the options for sensing valuable natural liquid such as honey. In this study, the purity of honey based on sugar content is investigated using the prism-based SPR technique. In order to create adulteration conditions, other sugar substitutes like fructose, glucose and sucrose are purposely added to pure honey (standard sample). It is based on its capability to detect bio molecular interaction whether biomolecules adsorption or desorption to the sensing surface [17]. The interaction leads to the mass density changes with high sensitivity on the sensing surface which in fact the refractive index is actually influenced by surface mass density too [18,19]. It is also believed that SPR is able to perform as it widely and successfully used to study and characterize various application fields in bio and chemical sensing [20]. Besides that, the advantage of SPR sensor as a non-destructive method makes the usage of the valuable and limited pure honey is able to minimize [21].

To the best of our knowledge, SPR is yet applied to detection of adulterated honey. Hence in this work, an attempt has been made to detect sugar (fructose, glucose and sucrose) adulterants in pure honey using SPR method. The footprints obtained will be able to provide useful information and parameters to develop a device that can foretell the sugar in adulterated honey or sugar contents in honey itself.

2. Theory

In SPR, a charge-density wave is corresponding to an electromagnetic wave. The wave propagates along the interface between two media with dielectric constants of opposite sign such as a metal and a dielectric. The propagation is characterized by the wave vector of evanescent field (K_{ev}) and wave vector of a surface plasmon (K_{sp}) as follows [22];

$$K_{sp} = \frac{\upsilon_0}{c} n_g \sin\theta \tag{1}$$

where υ_0 is the frequency of incident light, n_g is the refractive index of the dense medium (glass), θ is the angle of incident light and c is the speed of light in a vacuum [23], and;

$$K_{sp} = \frac{\upsilon_0}{c}\sqrt{\frac{\varepsilon_m n_s^2}{\varepsilon_m + n_s^2}} \tag{2}$$

where ε_m is the permittivity of metallic film and n_s is the refractive index of dielectric medium.

The field of surface plasmon wave is the transverse magnetic (TM) polarized and when the field vectors reach their maxima at the interface at a specific angle of incident, it decay evanescently into both media. This situation happens whenever evanescent wave of the incoming light is able to couple with the free oscillating electrons (plasmons) in the metal film corresponding to when $K_{sp} = K_{ev}$. Thus, the surface plasmon becomes resonantly excited in this situation.

3. Experimental setup

3.1. Materials and sample preparation

The authentic honey was purchased from commercial supplier and used without any purification. The standard sugar used were fructose and sucrose manufactured by Systerm Sdn Bhd while glucose was manufactured by Qrëc (Asia) Sdn Bhd. The prism employed in SPR setup was purchased from WTS Photonics Co., Ltd. For the glass slide (10 mm × 10 mm), it was acquired from Deckglaser. This size can be used directly on the custom liquid sample holder and ply on the prism sensing surface. Gold (Au) layer with a thickness of about 50 nm was sputter-deposited on the glass slide using a conventional sputtering system (Emitech K575X Turbo). Prior to that, the glass slide was cleaned using a precision dust removal equipment. In this work, only Au layer was used as the sensing surface without any intermediate layer [24].