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## Discrimination of Honey Adulteration Using Laser Technique

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### ABSTRACT

Honey has been a prime target for adulteration reasons of economic gain. Honey adulteration discriminated by chemical analysis or sophisticated devices which consume time, effort and money. The aim of this research is to use Laser Induced Fluorescence (LIF) technique, to get pure honey fingerprint and compared it with adulterated, heated and stored honeys. Fresh honey collected on June 2010., and adulterated by adding four different materials (glucose syrup, starch solution, distilled water, and molasses), with different concentrations (1,3,6,12 and 24%). Pure honey samples heated from 28 to 76 °C., and stored for two years. LIF spectra can be detect adulterated honey clearly by glucose or starch solution and distilled water or molasses starting from 3% and 1% concentration, respectively. Also, can through this technique discovered heated and as well as honey freshness. This technique proved that it is faster, more sensitive and less costly than other methods used in discriminated honey adulteration

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## INTRODUCTION

Honey is one of the oldest and best-loved sweetening agents for foods and, over the centuries, it has still retained a “natural” image (Aparna & Rajalakshmi, 1999). It is a popular natural sweetener that has high nutritional value and medicinal effects (Miraglio, 2001). For this importance, Florida law defines honey as, "... only the natural food product made by honeybees from the nectar of flowers or the saccharine exudation of plants, containing no additives"(Sanford, 2003). Most honey is produced by two species of bee, namely *Apis mellifera* (the so-called honey bee) and *Apis cerana* (Crane, 1975). The raw material for the production of “floral” honey is nectar, a dilute solution of sugars found in the nectaries of flowering plants.

Egypt owns about 17,516 apiaries (53 municipal apiaries and 17,463 foreign apiaries), include about 1,138,525 beehives (1,786 municipal beehives and 1,136,739 foreign beehives). These apiaries produce about 6.021 Gg (5.3 Mg and 6.015 Gg from municipal and foreign beehives, respectively), with total value about 126,759,000 LE (Economic affairs sector, 2011).

As a natural product of a relatively high price, honey has been a prime target for adulteration reasons of economic gain for a long time (Sivakesava & Irudayaraj, 2001a). Adulteration or the addition of foreign substances to honey is a reprehensible practice, which consists of incorporating sugar syrups into the genuine product (Cordella, *et al.*, 2005). For honey to be considered adulterated, the concentration of the adulterant should be at least 7% (Rossmann, *et al.*, 1992). Many foreign substances could be added to adulterate honey such as; molasses, starch solution, commercial glucose, sucrose, water, chalkm gelatins, inverted sugar and other substances (Abd El-Aal *et al.* 1993; Sharbt & Abdel- Fattah, 1994; Ruoff & Bogdanov, 2004; Bogdanov, 2010). The use of excessive heat for liquefaction or pasteurization has adverse effects on honey quality, i.e.(Ruoff & Bogdanov, 2004).

Detection of adulteration in honey is so difficult because of the large natural variability of honey, such as differences in species, maturity, environment, processing and storage technique (Sivakesava & Irudayaraj, 2001b). To ensure the quality and detect frauds, many methods have been tested for adulteration proof, but most of them are not capable to detect unequivocally adulteration (Bogdanov & Martin, 2002). The honey consumer establishes the quality of honey with eye, nose and mouth (Bogdanov, 2009). A list of the analytical methods that are used to prove the authenticity of honey had been reported (Anklam, 1998).

Adulterated honey was detected by changing in physicochemical, rheological and mechanical characteristics such as; refractive index, moisture content, T.S.S (total soluble solids), density, specific volume, capillary

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action, surface tension, pH value, viscosity and homogeneity (El-Biale & Sorour, 2011; Awad *et al.*, 2012). On the other hand, changes in the honey physical properties lead to changes in ultrasound velocity (Singh & Dwivedi, 1995). So, it can be used LPU (Low power ultrasound) to detect adulteration honey by measuring the high frequency dynamic shear rheology, viscosity and moisture content (Kulmyrzaev & McClements, 2000; Cereser & Laux, 2010.)

The capacity of honey to break the light is used for the refractometric determination of humidity (Bogdanov, 2009). Adulteration by addition low level (5 to 10%) of cane and beet sugar hydrolysates, high fructose corn syrup, can be screened microscopically (Kerkvliet *et al.* 1995; Kerkvliet & Meiger, 2000) or by carbohydrate chromatography method (Prodolliet & Hischenhuber, 1988) and verified by measuring the  $^{13}\text{C}/^{12}\text{C}$  isotopic ratio (White & Doner, 1978; Brookes *et al.* 1991; Rossmann *et al.* 1992; White, 1992; White *et al.* 1998). Recently this method has been further developed to include Site-Specific Natural Isotopic Fractionation (SNIF) measured by Nuclear Magnetic Resonance (Cotte *et al.* 2007).

A recent development is used global fingerprints such as chromatograms to detect the addition of beet sugar and determine full sugar profiles (sucrose- glucose- fructose ratios), by high – performance liquid chromatography (HPLC) or high – performance anion - exchange chromatography pulsed amperometric detection (HPAEC–PAD) (Cordella *et al.*, 2003; Elflein & Raezke, 2005; Cabanero *et al.* 2006). Meanwhile, the addition of high fructose corn syrup (HFCS) may be detected by detection of oligosaccharides naturally not present in honey through capillary gas (GC) (Low & South, 1995).

Recently, infrared spectroscopic methods using laser light in the near-infrared region has been applied for the detection of beet and cane sugar syrups sugar up to 4% to honey (Caceres *et al.* 2000., Sivakesava & Irudayaraj, 2001a., 2001b, 2001c; Paradkar and Irudayaraj, 2002; Kelly *et al.*, 2006a., 2006b). On the other hand, FT-Raman spectroscopy was investigated as spectroscopic fingerprint technique combined with some chemometric tools used as a simple, rapid, cost-effective, and non-destructive characteristics method for the discrimination of honey according to their source Pierna *et al.* (2011).

The main objective of this research is developing procedures for simple, rapid and non-destructive detection of adulterated honey. Specific objectives are: a) using LIF technique to obtain optical fingerprints for pure honey compared with adulterated, stored and heated honey, b) considered the first step to developed of an automated tool to discriminate adulterated honey, by identify the type and the concentration of additive materials using laser technique.

## MATERIAL AND METHODS

To achieve the objective of this research, the experiments were conducted in LASER physics laboratory at National Institute of LASER Enhanced Science (NILES), Cairo University from 2010 to 2012.

### **Honey:**

Pure honey (H) belong to clover plants (*Trifolium alexandrinanum*) were freshly collected from private foreign apiary beehives at Gharbia governorate, Egypt, on June 2010.

Honey properties are 18.1% moisture content measuring by digital refractometer, pH value of 3.76, determined using pH meter, density of  $1.407\text{ g cm}^{-3}$  measured by calibrated density bottle and dynamic viscosity, of 64 P measured at 30 °C using Brookfield digital rheometer, model DVIII Ultra and spindle No.HA-07 with container and spindle radius were 25 and 1.5 mm, respectively.

### **Substances fraud:**

Four different materials were added to adulterate pure honey namely; glucose syrup 20%(G), starch solution 3%(S), distilled water(W), and molasses (M).

### **Samples Preparation:**

Pure honey was adulterated by the addition substances fraud, at room temperature with different concentrations 1, 3, 6, 12 and 24%, each concentration were added separately. Adulterated samples were shaken well by electric vibrator for 5 min., to ensure good mixing. Sample volume was 50 ml. Five replicates from each samples were taken, each replicate were measured 3 times to avoid measurement error.

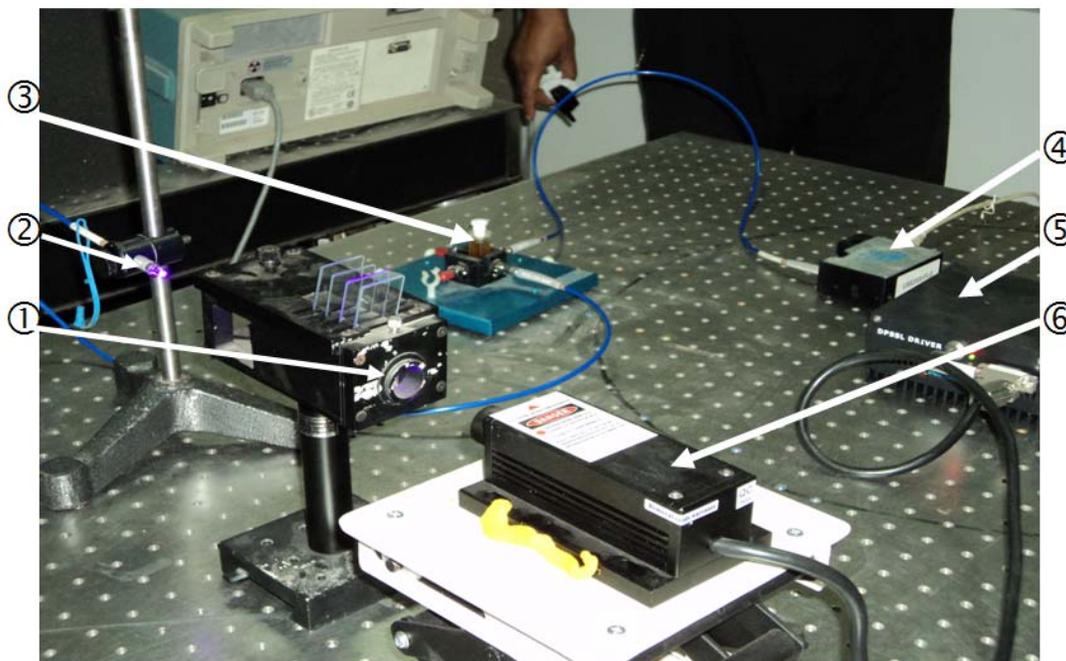
### **Heat Treatment And Freshness:**

Pure honey was heated in a water bath to increased honey temperatures to 28, 42, 54 and 76 °C.

The samples were kept at room temperature. A quantity of pure honey was stored for a period of two years with annual sampling for measurement. Samples taken two times during storing period after 12 (A) and 24 (B) month from harvesting day. All samples were stored in glass jars at room temperature ( $22 \pm 2$  °C).

**LIF Arrangement:**

The experimental set up of the equipment used throughout the present work for LIF measurements is shown in (Fig.1) The excitation light source was a continuous wave (CW) DPSS laser [Changchun new industries optoelectronics tech Co, Ltd. (CN)] with an average laser output power of 40 mW at a wavelengths of 405, 532 and 566 nm. The laser beam is focused into one end of an optical fiber while, the other end of the fiber delivers the beam onto one side of the quartz cuvette containing the sample. The emitted fluorescence is collected perpendicularly via another fiber used to deliver the fluorescent light to the spectrometer (USB2000 FLG, Ocean Optics, USA). Acquisition and analysis of the spectra obtained from the spectroscopic system is accomplished using the commercial Spectra Suit software (Ocean Optics, USA). All the spectra were further processed using computer software (Origin, Origin Lab. Corp., USA, Version 8).



**Fig. 1: LIF set up;** 1) Lenses. 2) Fiber optics (FO). 3) Quartz cuvette contains sample. 4) Detector. 5) Power supply. 6) Laser source.

## RESULTS AND DISCUSSION

This section deal & discuss basic three topics; 1) Appropriate LIF wavelength for the detection process. 2) Discriminate fraud materials and their concentrations. 3) Detected heated honey and freshness.

### 1. Appropriate LIF wavelength for the detection process:

Samples were exposed to laser beam at 405, 532 and 566 nm. The appropriate wavelength was 405 nm, which indicated fluorescence curve for each sample expect fraud materials. This result in harmony with that the honey fluoresce in range 250 – 420nm. (Ruoff *et al.* 2005, 2006).

### 2. Discriminate Fraud Materials And Its Concentrations:

The recorded LIF spectra at pure honey (H) and adulterated honey with glucose solution 20%(G), starch solution 3% (S), distilled water (W), and molasses (M), at concentrations 1, 3, 6, 12 and 24%, respectively., are displayed in (Fig. 2a, b, c & d)

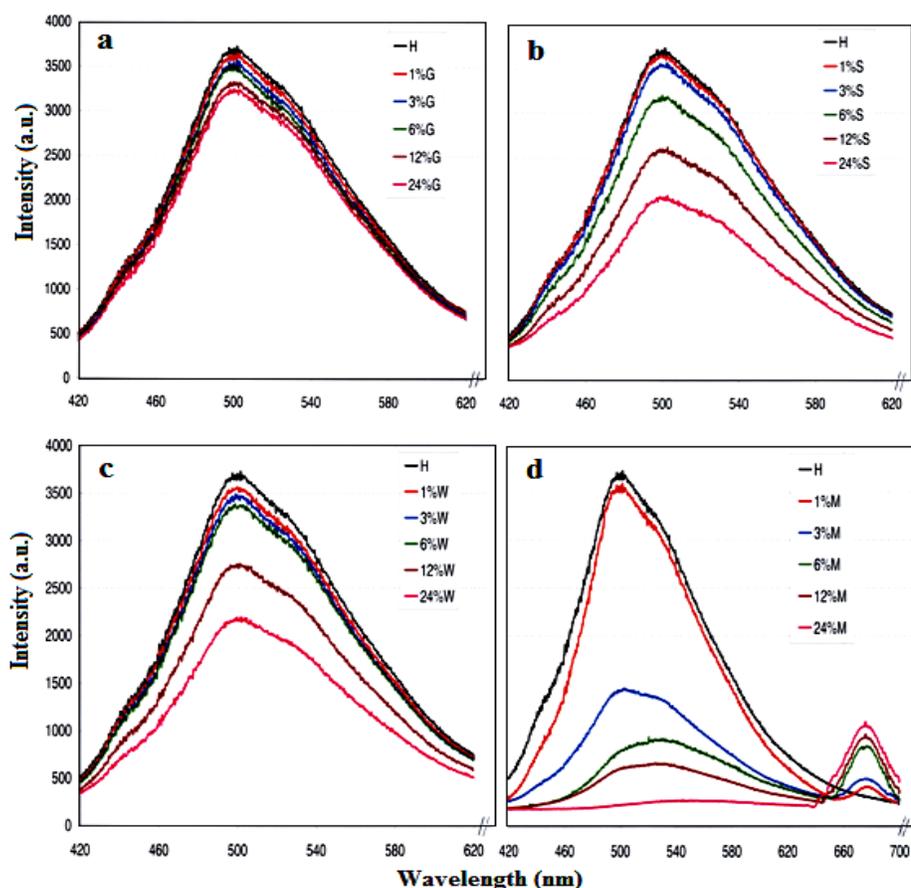
Excitation spectra were scanned from 420 to 620 nm (Fig. 2a, b & c ) and to 700 nm., (Fig. 2d). Pure honey peaking at about 500 nm., upon excited with 405nm.

Generally, it can be seen clearly that honey peak is decreasing by increasing the concentration of fraud, it can be distinguished between pure and adulterated honey easily and conveniently start from 3% concentration in all kinds of fraud (Fig.2a, b, c, & d). On the other hand, fraud can be discriminated with distilled water and molasses starting concentration of 1%.

Meanwhile, in honey adulterated with distilled water shows a clear decline of peak curve compared with pure honey curve (Fig.2c). Furthermore, increasing moisture content in pure honey more than 23% lead to honey fermentation (Bogdanov, 2010).

Adding any of glucose or starch or distilled water into pure honey considered to be dilution process. So, the overall shape of the curve has not changed with the exception of lower peak honey, that's where they have the same structural composition. (Fig. 2a, b, and c).

While, when adulterated with molasses occurs, a change in the form of trend. Beginning of a concentration of 1% molasses a small shoulder at 675 nm appears beside the main curve Fig.(2d). Where, increasing shoulder peak by increasing molasses concentration. The fluorescence comes from the transition from nonbonding, namely n electrons in the hetro - atom (O) of the functional group (C-O-C) called ether linkage, to the antibonding orbital 'sigma' (Haijin *et al.* 2005 ). This link is joins  $\alpha$ - glucose and  $\beta$  - fructose molecules to consisting sucrose molecule (non reducing oilgo sugar) which it represents 50% of molasses components. Meanwhile, Sucrose fluoresce in range 260– 465 nm. (Gabriela & Ionescu, 2003; Haijin *et al.* 2005 ).



**Fig. 2:** LIF spectra of adulterated honey with; a) Glucose solution 20%(G). b) Starch solution 3% (S). c) Distilled water (W). d) Molasses (M).

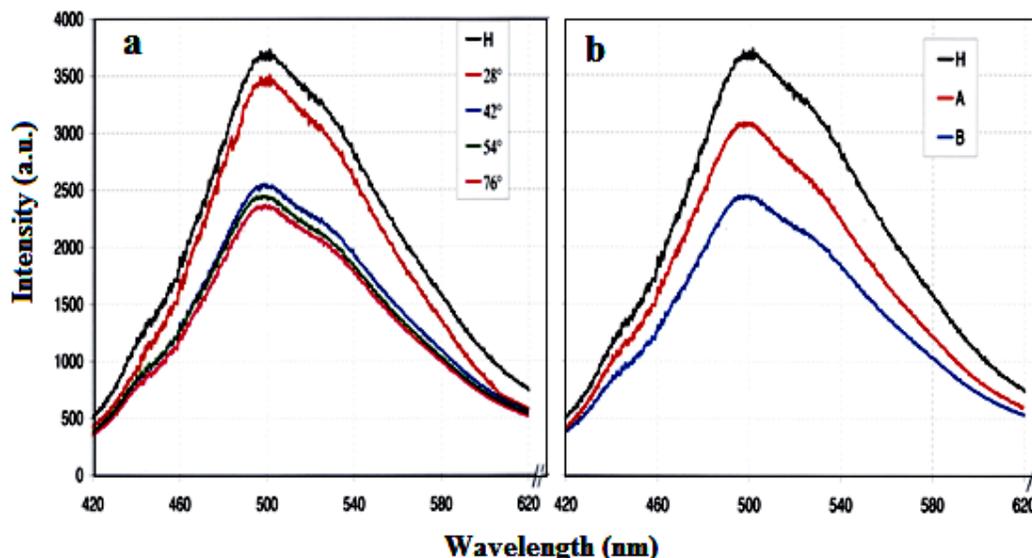
### 3. Detected Heated Honey And Freshness:

The fluorescence behavior from heated and stored honey shown in (Fig.3a & b). (Fig.3a), demonstrates the fluorescent behavior from pure honey and heated honey. It shows that, the heating honey leads to a reduction of honey peak. This decline slightly when heated from 22 to 28°C. While, the decline significantly and noticeably by increase heat from 28 to even 76°C.

Any large temperature fluctuation causes decay for honey. Heating honey up to 40 °C (104 °F) leads to destruct many of organic material and the most important enzyme diastase ( $\alpha$  -and  $\beta$ -amylase). It destroys invertase, and diastase activities (Melnyk, 2002; Oddo *et al.* 2004; Ruoff & Bogdanov, 2004). At the same time, heating leads to dehydrate sugar (fructose) and accumulate of HMF (Hydroxy methyl furfural) (Ruoff & Bogdanov, 2004; Yuriy *et al.* 2006; Andreia *et al.* 2011)

Whereas, the fluorescence behavior from stored honey are shown in (Fig.3b). It shows that, increasing storage period, lead to decreased the honey peak. Note that, the decline in the peak for (12 and 24 months), is almost equal, suggesting that the decline is going at a regular rate. The storage leads to inhibiting an enzyme diastase gradually and as well as, HMF is also slowly generated during storage (Arribas & Morales, 2010).

The results could be in harmony with storage for a period of one year leads to a declining in the number diastase No., from 14.6 to 10.7 and increased HMF from 3.8 to 19.1 mg kg<sup>-1</sup> (Yilamaz, & Kufrevioglu, 2001).



**Fig. 3: LIF spectra of adulterated honey with; a) Heated honey. b) Stored honey.**

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#### Conclusion:

This paper showed the possibility of discriminated honey adulteration by LIF spectra. Where, it can detect honey adulteration by glucose or starch solution and distilled water or molasses starting from 3% and 1% concentration, respectively clearly. Also, can through this technique discovered heated and as well as honey freshness. This technique proved that it is faster, more sensitive and less costly than other methods used in discriminated honey adulteration

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