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Low cost smart phone diagnostics for food using paper-based colorimetric sensor arrays

Yu Chen [a], Guoqing Fu [a], Yael Zilberman [a], Weitong Ruan [a], Shideh Kabiri Ameri [a], Yu Shrike Zhang [b], Eric Miller [a], Sameer R. Sonkusale [a, *]

[a] Electrical and Computer Engineering Department, Tufts University, Medford, MA 02155, USA
[b] Division of Engineering in Medicine, Department of Medicine, Brigham and Women's Hospital, Harvard Medical School, Cambridge, MA 02139, USA

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ABSTRACT

There is a need for an accurate end-of-life indicator for packaged food (meat, seafood, dairy food etc.) beyond a simple "best use by" date on the food package. In this work, we propose a low cost solution by repurposing the food's barcode as a colorimetric sensor array to monitor food condition. A smart phone camera is used to read color information from the sensor barcode for quantitative estimate of the food aging and quality. The sensor is based on cross-reactive vapor sensitive dyes encapsulated in resin microbeads, which are impregnated onto a low cost paper substrate in a barcode pattern. The entire sensor platform is validated by accurately monitoring chicken aging and eventual spoilage under different temperature conditions. The proposed food diagnostics platform has the potential to reduce food waste and eliminate food-borne illness.

1. Introduction

Generally, food quality is monitored during the food supply chain by qualified food inspectors using relatively expensive equipment. Some common techniques for food quality monitoring are gas chromatography (Monti et al., 2005), spectroscopy (Karoui, Downey, & Blecker, 2010), electromagnetic interrogation (Tao et al., 2012), ultrasound interrogation (Awad, Moharram, Shaltout, Asker, & Youssef, 2012) and chemical sensing using electronic nose (Loutfi, Coradeschi, Mani, Shankar, & Rayappan, 2015). However, regardless of the fine accuracy of these systems, they are neither cost effective nor user friendly for direct use by consumers to monitor the food quality at point of consumption. Consumers usually judge whether food is acceptable for consumption by looking at the expiration date, which is sub-optimal and has resulted in healthy food being discarded or unhealthy food being consumed (Wilson, Rickard, Saputo, & Ho, 2015). If one could implement a low cost sensor platform that could objectively provide the consumer with the information on food condition, it would be a significant step in avoiding global food wastage and in preventing food borne illness.

In recent years, several research groups have successfully employed colorimetric sensor arrays based on various optical dyes (Lin, Jang, & Suslick, 2011) (Soga, Jimbo, Suzuki, & Citterio, 2013) (Buryak & Severin, 2005). These sensor arrays have shown sensitivity to sub-ppm levels of target gases in their environment (Feng, Musto, Kemling, Lim, & Suslick, 2010). The most commonly used optical dyes for colorimetric sensors are metalloporphyrins, Brønsted acid/base dyes, solvatochromic dyes, and redox indicators (Lim, Kemling, Feng, & Suslick, 2009). They have been utilized for sensing volatile organic compounds (Janzen, Ponder, Bailey, Ingison, & Suslick, 2006), environmental pollutants (Kim, Ren, Kim, & Yoon, 2012), and toxic industrial chemicals (Feng et al., 2010). Interestingly, the volatile gases that the foods emanate can be used to estimate its freshness and quality. This has been the basis of many sensor platforms utilized for food sensing. A variety of optical dyes, including pH indicators, Lewis acid and natural dyes has been incorporated into inorganic support materials to monitor the gas products of meat spoilage (Li & Suslick, 2016). Similar ideas have been used for monitoring the freshness of pork (Huang et al., 2014), fish (Morsy et al., 2016) and chicken (Khulal, Zhao, Hu, & Chen, 2016) under the package atmosphere. While these

* Corresponding author.
E-mail address: sameer@ece.tufts.edu (S.R. Sonkusale).

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colorimetric sensor arrays have shown promise for food monitoring, they are still not amenable for direct consumer use. This is due to the need for expensive and bulky accessory to sample and deliver food-related gases to sensors, and of the optical readout instrumentation. Moreover the cost of fabrication in making some of these sensors may be cost prohibitive for many food items. In this paper, we demonstrate a low cost easy to use sensor platform using colorimetric sensor arrays with mobile phone camera based readout for direct consumer use at point of consumption.

There are plenty of advantages of using mobile phones for optical readout. First, in some of the less developed regions of the world, where there is little or no electricity, there are more mobile phones than landlines. Many of these phones have cameras with sufficient color and image resolution for scientific imaging. In fact, smart phone for medical diagnostics have already been proposed. For example, smart phone has been successfully used in a number of chemical and biological sensing applications (Ozcan, 2014) (Wei et al., 2013) (Zhu et al., 2013). Some examples include using smart phone as sensor read out of pattern change on a paper-based blood typing sensor (Guan et al., 2014) and of colorimetric detection of pH change in sweat and saliva (Oncescu, O'Dell, & Erickson, 2013). Smart phone implemented fluorometer has been successfully used for detecting specific nucleic acid sequence in a liquid sample (Yu, Tan, & Cunningham, 2014). It is shown that the color information captured by smart phone can be represented using various RGB color models, such as HSL or CIE. Some smart phone application algorithm has also been developed to directly quantify color information in the smart phone interface (Yetisen, Martinez-Hurtado, Garcia-Melendrez, da Cruz Vasconcellos, & Lowe, 2014). It has been proven that the smart phone-based colorimetric detection scheme can achieve the same accuracy as a benchtop spectrometer (Shen, Hagen, & Papautsky, 2012). The successful use of smart phone shows promise for realization of low cost sensor platforms leveraging existing consumer devices like smartphones as a substitute for an otherwise expensive scientific instrumentation.

Combining the advantages of optical dyes for gas sensing and smart phone detection, we present a disposable paper-based colorimetric geometric barcode sensor for monitoring food quality. The sensor is fabricated by stamping dye-encapsulated silica beads that are pH and volatile organic compounds (VOCs) responsive on a paper substrate in geometric shapes resembling a barcode. The as-fabricated sensor can be directly attached to the surface of the food (e.g. meat) or onto the inner lining of the food package to monitor its condition. The sensor then changes its color based on the volatiles emanating from the food, which is trapped inside the sealed package. For readout, any smart phone with a camera is used to capture the images of the sensing barcode, and a builtin app which directly measures the color change in this sensing barcode using image processing to generate quantitative results. Compared to the existing work on food monitoring, our sensor platform has several advantages. First, the sensor fabrication process is low cost and highly reproducible; Second, compared to electrochemical sensing based approach, utilizing optical dyes as sensor provides high sensitivity and rapid response as shown in prior work on using such dyes in gas sensing (Rakow & Suslick, 2000) (Lin et al., 2011). Moreover optical sensing provides natural wireless means for readout. Our approach aims to incorporate colorimetric dye sensing arrays into the product barcode or QR code for simultaneously obtaining food UPC code and condition/quality information. Some existing bio-barcode applications include immobilizing small DNA molecules onto gold nanoparticles to form a barcode for pathogen detection (Zhang, Huarng, & Alocilja, 2010) or converting photoluminescence spectrum of nanoporous anodic alumina into barcode (Santos et al., 2012). The more popular QR code is able to store more information and is highly compatible with smartphone readout. Compared with monochrome barcode, colorful QR code has the advantage of higher data storage and lowering the readout error rate (Ramya & Jayasheela, 2014). Using optical dyes to make such colorful QR code offers significant promise for sensing and tracking of food. The convenience of using smartphone for scientific readout makes it amenable for use by consumers at home. Third, in terms of fabrication, we employ the resin encapsulation of the colorimetric optical dyes, which prevents food contamination from dye leakage. Finally, we can apply machine learning approaches on colorimetric data to provide accurate assessment of food quality. For proof of principle, we utilized the principal component analysis (PCA) on the quantitative data from packaged chicken meat where we show that we can not only monitor aging status of the chicken each day, but also at an interval of every hour, making this a very sensitive platform to estimate food aging and quality.

2. Materials and methods

2.1. Preparation of sensing dyes

Nile red, Zinc Tetraphenylporphyrin (Zn-TPP) and Methyl red (Sigma Aldrich) were used for sensing emanating gases from the chicken meat. The procedures for preparing Methyl red and Nile red microbeads solutions are similar: 25 mg of each dye powder was first dissolved in 5 mL of de-ionized water, and this mixture was stirred using a magnetic stirrer for 20 min before being poured into 256 mg of anion exchange resin microbeads (Sigma Aldrich). The resulting mixture was stirred again for 2 h at room temperature to fill the resin microbeads with optical dyes (Yu Chen, Zilberman, Mostafalu, & Sonkusale, 2015) (Y. Chen et al., 2016). The method of preparing Zn-TPP beads solution is described in detail in our previous work (Zilberman, Chen, & Sonkusale, 2014). Briefly, 25.6 mg of Zn-TPP powder was first dissolved in 3 mL of toluene and stirred using a magnetic stirrer for 2 h to ensure good dissolution. The dye solution was poured into 256 mg cation exchange resin microbeads. The resulting mixture was again stirred for 2 h at room temperature to fill the microbeads with optical dyes. The as-prepared Zn-TPP micro-beads solution was left in the fume hood over-night for toluene to evaporate. 3 mL of de-ionized water is added into the dried Zn-TPP micro-beads.

2.2. Sensor fabrication

The sensor fabrication process is described conceptually in Fig. 1. As shown in Fig. 1a, the microbeads containing various optical dyes were prepared based on a recipe discussed in the previous section. Each category of optical dye-loaded beads is stored in its own container and is ready for use. A polymer mold with triangle, square and circle grooves was made through laser cutting (Fig. 1b). These shapes were chosen as a geometric substitute for QR code to show our capability to pattern complex geometry. Moreover the shapes enable one to distinguish the different sensing elements using shape-based encoding since each dye correspond to a unique shape pattern and size. The de-gassed PDMS was poured into the as-fabricated polymer mold and cured in the oven under 80 °C for 5 h. After that, the cured stamp was peeled off the mold (Fig. 1c). In Fig. 1d, three types of dye-contained micro-beads were drop-casted on to the triangle, square and circle patterns of the stamp using a micro-pipette (20 μL), respectively. The dye-loaded microbeads organized in the geometric pattern on the stamp is simply attached to the double-sided tape (Fig. 1e), to transfer them in their geometric pattern onto the tape (Fig. 1f). This micro-beads-attached tape was then attached to filter paper (as protective covering and a barrier) to complete device fabrication. The image of as-fabricated