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# Fabrication of paper-based devices by lacquer spraying method for the determination of nickel (II) ion in waste water



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## ABSTRACT

A spraying method with lacquer was developed for the fabrication of paper-based devices. A patterned iron mask was initially placed on a filter paper and held tightly attached by a magnetic plate placed on the opposite side. After that, acrylic lacquer was sprayed on the filter paper to create a hydrophobic area while the hydrophilic area was protected with the iron mask. The optimal conditions for the fabrication of this device were studied including lacquer type and particle retention efficiency of filter paper. Gloss spray lacquer and filter paper No. 4 were chosen as optimal lacquer type and particle retention efficiency of filter paper, respectively. To evaluate its efficiency, the paper-based devices were used to determine nickel using electrochemical detection. Cu-enhancer solution was employed to increase sensitivity of nickel determination with the optimal concentration of 4.5 ppm. Under the optimal conditions, linear range was observed in the range of 1–50 ppm with a coefficient of determination of 0.9971. The limit of detection (LOD) and the limit of quantitation (LOQ) were found to be 0.5 and 1.97 ppm, respectively. Moreover, these paper-based devices coupled with electrochemical detection were applied to determine nickel in waste water of a jewelry factory and compared to those obtained with inductively coupled plasma optical emission spectrometry (ICP-OES). The results indicated that there were no significant variations between this proposed method ( $4.15 \pm 0.043$  ppm) and the ICP-OES method ( $4.06 \pm 0.013$  ppm). Therefore, this spraying method was found to be an excellent alternative for the fabrication of paper-based devices due to its ease of use, affordability and simplicity.

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## 1. Introduction

Paper-based devices have the potential to be good alternative analytical devices for healthcare related applications because they are portable, easy to use, have a low sample volume requirement and provide rapid analysis [1–3]. Paper-based device is a device made of paper in which the main component is cellulose fiber. Paper is a low-cost material that is readily available and easy to manipulate. Therefore, paper-based devices have been widely popular and have many potential benefits in fields as diverse as environmental monitoring and clinical research. Currently, paper-based devices have become an interesting technology for research units, resulting in the development of method for the fabrication of paper-based devices. A lot of methods for fabricating the pattern on paper have been proposed, including photolithography [4–11], polydimethylsiloxane (PDMS) plotting [12], inkjet printing [13],

cutting [14], plasma etching [15], wax printing [16–18], wax screen-printing [19], and wax dipping [20]. Each fabrication method has its own advantages and limitations that are similar and/or different. The first reported method was based on photolithography. This method can provide a narrow barrier line between the hydrophilic and hydrophobic areas of approximately 200  $\mu\text{m}$  width [6]. However, this method involves the use of organic solvents, expensive photoresists and photolithography apparatus. Moreover, the fabrication process involves many complicated processes. The PDMS plotting method also overcomes the problems of physical inflexibility of devices using photolithography [11]. This method does not use expensive photoresists, organic solvent and photolithography instrument. Unfortunately, this method needs a customized plotter [10]. The inkjet printing method involves the use of organic solvent to print onto the polymer-soaked paper by inkjet printer for creating a hydrophilic area on the paper. Plasma etching is a method to detract sizing agent [21] coating on the paper using plasma treatment. However, paper-based devices created by photolithography, inkjet printing and plasma etching methods all still require organic solvents and polymers to create hydrophilic areas. In the cutting method, a knife plotter is used to cut paper to create

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a pattern of microfluidic channels. However, this method requires the use of tape to define the paper pattern, which is difficult in implementation. The wax printing method utilizes a commercially available wax printer for the fabrication of paper-based devices. This method is easy and fast to generate a patterned paper using a wax printer. However, this method has a shallower barrier than photolithography ( $\sim 850 \mu\text{m}$  of minimal barrier) because the spread of the wax is difficult to handle when wax was melted on hotplate. So, careful impregnation of wax must be regarded before creation of the pattern in this method. The wax screen-printing method is similar to the wax printing method by using a commercial wax for the fabrication of paper-based devices, but wax screen-printing does not require wax printer for fabrication. This method can create a pattern by using screen-printed block instead of the commercial printer. The advantages of the wax screen-printing method are low-cost, simple, and rapidness. Nevertheless, using wax screen-printing it is difficult to produce the exact designed pattern with high barrier due to the spread of wax. Recently, a new fabrication method for creating paper-based devices has been reported as wax dipping. Wax dipping does not demand expensive equipments and organic solvents. However, the hydrophobic areas generated by the wax printing, wax screen-printing and wax dipping methods still used the hot plate for melting wax. Moreover, limitations of these previous methods by wax are wax spreading before creating the pattern and the trained personnel for using and maintaining the instruments. To overcome these limitations, a simple, rapid and low-cost fabrication method, that also provides several advantages, needs to be developed. A spraying method with lacquer was therefore developed for the fabrication of paper-based devices.

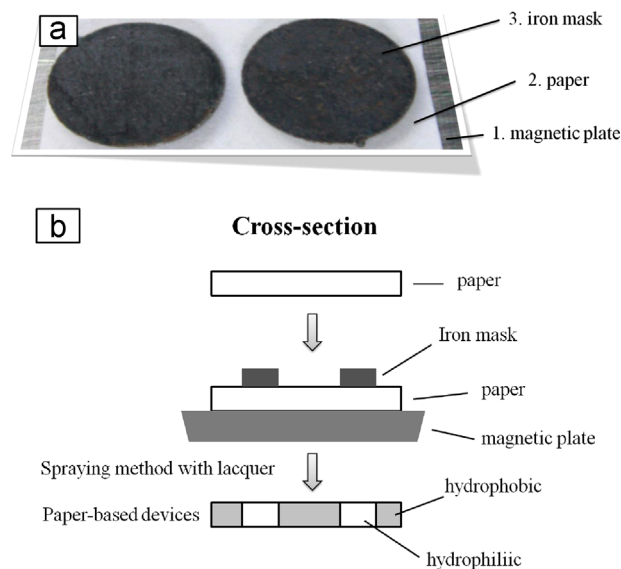
Acrylic lacquer is made of acrylic resin. One of main characteristic features of acrylic resin is high transparency. Acrylic lacquer is a polymer (resin) generated through chemical reaction by applying polymerization. The advantages of acrylic resin are water resistance, good adhesion and fast drying [22].

The objective of this work is to present a new concept of spraying method with lacquer for the fabrication of paper-based devices. To evaluate the efficiency of this developed method, paper-based devices with electrochemical detection were used to determine nickel in waste water of a jewelry factory using the differential pulse anodic stripping voltammetry (DPASV) and copper-enhancing solution was used for increasing the sensitivity of nickel determination.

## 2. Materials and methods

### 2.1. Materials and chemicals

Whatman No. 1 (11  $\mu\text{m}$  porosity) and No. 4 (20–25  $\mu\text{m}$  porosity) filter papers were purchased from Cole-Parmer (Vernon Hills, IL). A patterned iron mask (1 mm thick, manufactured from steel plate) was made-to-order by a laser cutting shop in Bangkok. Magnetic plate and acrylic lacquer Leyland<sup>®</sup> manufactured by Nakoya Paint (Thailand) Co., Ltd. were purchased from a local area shop in Bangkok. Carbon ink was purchased from Gwent group (Torfaen, UK). Silver chloride ink (Electrodag 7019) was obtained from Acheson Colloids Company (Port Huron, MI). Electrochemical measurements were performed using a potentiostat (Autolab PGSTAT 30). All solutions were prepared in  $18 \text{ M}\Omega \text{ cm}^{-1}$  resistance deionized water (obtained from a Millipore Milli-Q purification system). A standard solution CertiPUR<sup>®</sup>,  $1000 \text{ mg L}^{-1} \text{ Ni}(\text{NO}_3)_2$  in  $\text{HNO}_3$  2–3% was purchased from Merck and was used as the stock solution. Sodium chloride (NaCl) (99.5%) (Merck) and copper sulfate ( $\text{CuSO}_4$ ) (99%) (BDH) were used as received.



**Fig. 1.** Schematic representation of the procedure for the fabrication of the paper-based devices by the lacquer spraying method (a) in top view and (b) in cross-section view.

### 2.2. Spraying method with lacquer for fabrication of pattern on paper

The spraying method with lacquer was used to fabricate the pattern on a filter paper. The iron mask (Fig. 1a), which was designed by an Adobe Illustrator and manufactured using the laser cutting technique, was used to create cover pattern on the paper. Hydrophobic area was created by the spraying method. To fabricate paper-based devices (Fig. 1b), the paper was first put on a magnetic plate. Next, the iron mask was placed on the other side of the paper and it was temporarily attached by means of magnetic force with a magnetic plate placed on the backside of the paper. Then, the paper was sprayed with lacquer creating the hydrophobic barrier around the iron mask. After that, the paper was air-dried and the iron mask was removed from the paper. Finally, the image of hydrophobic and hydrophilic areas of the pattern on the paper were characterized using an optical microscope and scanning electron microscopy (SEM)

### 2.3. Preparation of electrochemical detector for paper-based devices.

The screen-printed electrodes were prepared in-house. The three electrodes were fabricated using the screen-printing method. The carbon ink was used as the working electrode (WE) and the counter electrode (CE) and the silver/silver chloride ink was used as the reference electrode (RE) and conductive pads. All electrodes were screened on the patterned paper and were cured in the oven at  $65 \text{ }^\circ\text{C}$  for 30 min. The paper-based devices coupled with electrochemical detection are shown in Fig. 2.

### 2.4. Applicability of paper-based devices for the determination of nickel in waste water sample of a jewelry factory

A waste water sample from a jewelry factory was obtained from the Gem and Jewelry Institute of Thailand (Public Organization). The jewelry waste sample was prepared by filtration of 20 mL of waste water with Whatman No. 1 filter paper. Then the sample was mixed with 20 mL of  $\text{HNO}_3$  and the solution was heated at  $200 \text{ }^\circ\text{C}$  to evaporate the solvent. After that, the sample was made up with 20 mL of Milli-Q water and was heated at  $100 \text{ }^\circ\text{C}$  until dryness (repeated three times). Next, the sample was rinsed with

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