



## A paper microfluidic fuel cell powered by urea

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### HIGHLIGHTS

- The first paper microfluidic fuel cell powered by urea is presented herein.
- This paper fuel cell produces a maximum  $0.91 \text{ mW cm}^{-2}$  power and  $4.3 \text{ mA cm}^{-2}$  current.
- This paper fuel cell operates on 0.3 M urea, the concentration found in urine.
- The fuel cell with 0.3 M urea powers a pregnancy test for over 20 min.

### ARTICLE INFO

#### Keywords:

Portable power  
Galvanic cell  
Urea fuel cell  
Urine analysis  
Pregnancy test

### ABSTRACT

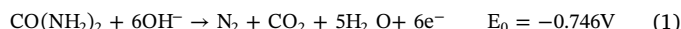
A paper microfluidic fuel cell was developed that is powered by urea and compares favorably to recent literature reports of paper microfluidic fuel cells. During fuel cell testing, we found the maximum power density to be as high as  $0.91 \text{ mW cm}^{-2}$  and the maximum current density to be as high as  $4.3 \text{ mA cm}^{-2}$  at room temperature using 0.3 M urea as fuel. Under continuous reactant flow, the fuel cell voltage is stable for at least 1 h. Under stopped flow conditions, the voltage was stable for up to 20 min. When 3 fuel cells (using Ni anode) were connected in series, sufficient energy was produced to power the LCD screen of a paper-based, over-the-counter pregnancy test for at least 30 min. Since this fuel cell is constructed on paper, it does not require continuous flow of reactants as a micro-channeled microfluidic fuel cell would. Hence, it could be possible with this chemistry to integrate energy production with urinalysis on paper since human urine contains both urea (fuel) and hormones (analyte) to provide power and analyte simultaneously.

### 1. Introduction

In 2002, the first microfluidic fuel cell (MFC) was developed as a membrane-less device that relied on co-laminar flow to prevent mixing between the fuel and oxidant streams in place of a physical barrier such as a cation exchange membrane [1–3]. Without the technological restrictions of a membrane, several fuels at various pH conditions have been studied, including hydrogen, methanol, formate, formic acid, hydrogen peroxide, and vanadium [4–12]. The co-laminar flow principle (found in microchannels and on paper) prevents complete mixing of two streams but permits a small interdiffusion zone that allows ions to complete the charge migration required to produce power [13].

Recently, Zhang et al. successfully developed a membraneless urea microfluidic fuel cell (UMFC) that extracts energy from urea using a Ni-supported-carbon-nanotube-coated sponge (Ni/CNT@Sponge) as the anode catalyst and air as an oxidant at the cathode [14]. Since urea hydrolyzes to ammonia in aqueous solutions, urea is not compatible with cation exchange membranes such as Nafion that are employed in many direct liquid fuel cells. Hence, its efficiency is expected to be

higher in a microfluidic environment that is not limited by membrane technology. The anode reaction for the oxidation of urea in alkaline conditions is:



Zhang et al. paired this oxidation reaction with the reduction of oxygen in an acidic environment ( $E_0 = 1.229 \text{ V}$ ) to design the fuel cell to have a theoretical open circuit voltage (OCV) of 1.975 V. In practice, overpotential losses reduced the OCV to 0.92 V at 20 °C when the combination of 3 M urea and 3 M KOH were used as the anolyte and 1.5 M  $\text{H}_2\text{SO}_4$  as the catholyte. Peak power density and maximum current density of  $3.9 \text{ mW cm}^{-2}$  and  $23 \text{ mA cm}^{-2}$  were achieved, respectively, under the same fuel and oxidation conditions [14].

A very significant limitation to a microfluidic fuel cell is that it requires continuous pumping to maintain continuous flow at the anode and cathode to prevent reactant mixing; this pumping results in parasitic losses that decrease the overall power output of the fuel cell. In addition to the work of Zhang et al., there have been a few significant reports of non-microfluidic operation of a direct urea fuel cell.

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Lan et al. first reported the operation of a direct urea/urine fuel cell (DUFC) that consisted of an alkaline anion exchange membrane (AEM). This fuel cell employed a nano-sized nickel anode catalyst and 1 M urea as fuel. An OCV of 0.65 V was achieved, and maximum power density of  $1.8 \text{ mW cm}^{-2}$  and maximum current density of  $6 \text{ mA cm}^{-2}$  were realized at  $20^\circ\text{C}$  [15,16]. This motivated the work of Xu in which NiCo/C catalysts were investigated for DUFC performance, and 10% Co to 90% Ni was found to be the most efficient catalyst ratio, producing a maximum power density of  $1.6 \text{ mW cm}^{-2}$  when 0.3 M urea was used as fuel at elevated temperature.

Guo et al. designed a direct urea-hydrogen peroxide fuel cell (DUHPFC) that involved nickel-cobalt nanowire arrays as the anodic catalyst, combination of 0.33 M Urea and 9 M KOH as the anolyte and combination of  $\text{H}_2\text{SO}_4$  and  $\text{H}_2\text{O}_2$  as the catholyte. Peak power density of  $7.4 \text{ mW cm}^{-2}$  and an OCV of 0.92 V were achieved at room temperature using  $\text{Co}_{10}\text{Ni}_{90}$  anode catalyst [17]. They also investigated 3D porous Ni-Co film on nickel foam catalysts of several cobalt/nickel ratios, finding that the DUHPFC performance was even greater on Ni-Co/Ni foam. This fuel cell produced an OCV of 0.83 V and a peak power density as high as  $17.4 \text{ mW cm}^{-2}$  when operating using 0.5 M urea as fuel and 7.0 M KOH as electrolyte. They also demonstrated that human urine could be utilized to produce an OCV of 0.80 V and a maximum power density of  $7.5 \text{ mW cm}^{-2}$  [18].

Several other studies have been performed on the electrochemical oxidation of urea but *without fuel cell performance reported*. The commonality in these studies is the use of nickel catalyst, but several different admetsals have been analyzed with the Ni including: Co, Zn, Sn, Cr, Pt [19–27].

In this work we transfer the electrochemical principles governing the urea fuel cell to a paper microfluidic environment for the first time to our knowledge. This paper environment is advantageous to the direct integration of power with sensing and does not demand continuous flow to maintain separation of anolyte and catholyte. Esquivel et al., and Copenhaver et al., demonstrated the first paper microfluidic fuel cells powered by methanol and formate, respectively, and showed that such fuel cells have potential to be used for point-of-care testing diagnostics by storing the fuel materials directly on the paper strip being used for the test [11,28]. In contrast, the urea fuel cell discussed in this present work utilizes the fuel (urea) already present in human urine to reduce the quantity of materials that must be pre-deposited on the paper strip.

## 2. Experimental

Whatman 3 filter paper was used to construct microfluidic, Y-shaped fuel cells having a channel length of 4.5 cm and channel width of 1.8 cm in the design of previous work with paper microfluidic fuel cells; the design principle is shown in Fig. 1 [11,29]. The Y-shaped design transports reactants to the co-laminar flow region of the paper where the anolyte and catholyte streams flow next to each other in 0.9 cm channels with a small enough quantity of mixing that the anolyte does not contact the cathode and the catholyte does not contact the anode.

The electrodes consisted of four small dots at anode and four at cathode, each dot having diameter slightly less than 0.4 cm and painted directly on the paper using a plastic nail-dotting tool. Anode dots were Pt (high surface area, Alfa Aesar, 0.3 mg per dot) or Ni metal (powder, Fisher, 0.3 mg per dot) mixed with Nafion binder and water. Cathode dots were carbon (colloidal graphite, Ted Pella, 1.2 mg per dot). The size and shape of the circular dots has been optimized to prevent inhibition of flow of the anolyte and catholyte that occurs when larger, square electrodes are painted. Scalable increases in power were not found when more dots were applied.

The fuel cell behaviour was monitored using a Princeton Applied Research 263 A potentiostat with a two electrode configuration having the anode and cathode connected using brass current collectors and

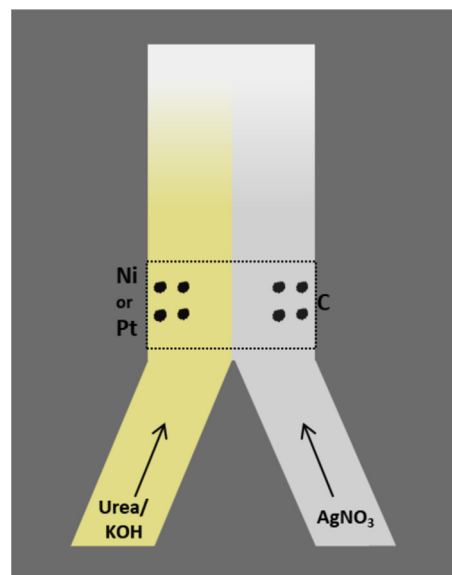


Fig. 1. The design principle of the paper microfluidic fuel cell powered by urea is shown as a paper device that promotes co-laminar flow of urea/KOH over Pt or Ni electrodes and silver nitrate [13] over carbon electrodes.

alligator clips. Surface area was determined from the geometric area of the total number of dots on each electrode; the dots were slightly less than 0.4 cm diameter. Voltage current (VI) measurements were taken with 2 s, 20 mV steps from open circuit to 10 mV. Constant current measurements were taken at  $0.25 \text{ mA cm}^{-2}$  for 1 h under continuous flow of reactant using an absorbent pad to collect the waste stream from the top of the Y-shaped fuel cell. The device was also analysed for constant current under stopped flow conditions, whereby the Y-shaped channels were removed from the reactants after the paper was saturated with reactants; the measurement was taken until device failure due to consumed reactants. An electronic pregnancy tester was also used (e.p.t. early pregnancy test) to determine the utility of the fuel cell in a practical application.

## 3. Results and discussion

### 3.1. Working principle

The fuel cell design shown in Fig. 1 allows co-laminar flow of the 2 reactant streams that prevents mixing that would deactivate the cell. A co-laminar flow device utilizing microchannels requires continuous pumping to maintain the co-laminar flow and prevent mixing. In contrast, our device maintains separation of the reactants even after they cease flowing when the paper is removed from the reactant source vials [11–13]. Hence, this device can be operated under a stopped flow condition similar to that of a water-based test where reactants are provided for a few seconds or minutes with capillary action used to move the reactants toward the location of analysis. If the same fluid that is being analysed also contains reactants for a fuel cell or galvanic cell, then both analysis and energy production can occur on the same strip of paper simultaneously; this would eliminate the need for the device battery which contains materials that are not consumed and are hazardous to the environment when discarded. Recent work has highlighted the opportunities that paper microfluidics provides for analytical sensing and integrated power production using fuels such as methanol, formate, and hydrogen, as well as metal redox reactions [11–13,28,30–33].

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