Improved low-cost, non-hazardous, all-iron cell for the developing world

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HIGHLIGHTS

- Low-cost non-hazardous all-iron cell is improved using commercial materials.
- Maximum discharge power of 40 mW cm^{-2} was observed.
- Ferric iron utilization of 88% is achieved.
- Demonstrated prototype LED lighting product that consumes inexpensive iron materials.

ARTICLE INFO

ABSTRACT

A low-cost, non-hazardous personal-power system based on an aqueous all-iron electrochemical cell is demonstrated. The system is intended to be assembled and operated by developing-world households that lack sufficient access to electricity, thereby enabling LED lighting or mobile phone charging on demand. Lab-scale hardware is used to assess the performance of individual cell components. It is found that coffee filter paper is an effective low-cost separator. Carbon felt is a low-cost electrode material, and its performance and wetting by the electrolyte solution is greatly improved by pre-treatment with sulfuric acid. The carbon felt does not degrade after a week of daily use. By using these components, performance of the system is significantly improved over the previous baseline, with power density more than doubling to 40 mW cm^{-2}, and iron utilization improving from 78% to 88%. The operating cost is estimated to be less than US$0.03 per mobile phone charge. Based on the lab-scale results, a stand-alone prototype consumer product is designed, fabricated, and tested. It successfully provides 2.5 h of LED illumination while consuming 200 mL of electrolyte solution via gravity feed. We anticipate these results will enable deployment of this innovative system to energy-impoverished individuals in the developing world.

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1. Introduction

Insufficient access to modern electrical power is prevalent throughout the developing world [1]. 500 million households rely on kerosene-lamp lighting, which provides low-quality light; it is unsafe due to indoor air pollution, kerosene ingestion, and fire hazard [2], and is expensive with typical household fuel costs around $4 per month [3]. Cellphone ownership is common througho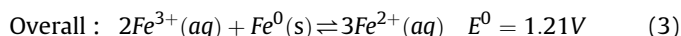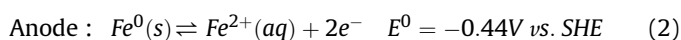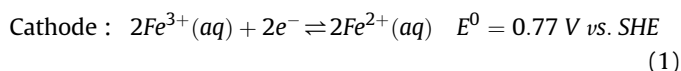ut the developing world, although access to electricity to charge the phones is scarce. In sub-Saharan Africa, roughly 80% of adults own cellphones [4], yet electrification rates are only 59% of urban and 17% of rural households [5]. Charging mobile phones at pay-per-charge kiosk businesses costs about $0.25 per charge.

To address this need, we previously introduced a concept for a low-cost, non-hazardous iron-based battery in which the active materials are assembled by the end-user, and can be spilled on the ground after use without impacting environmental or human health [6]. The vision is a compact, low-cost power source that allows end-users to produce small amounts of power suitable for LED lighting and phone charging in their home, on demand. The user will initially purchase a durable product containing a cell housing, electrical circuitry, LED lamp and USB receptacle (for

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phone charging). Fig. 1 shows the configuration of the active electrochemical components, including a porous carbon (+) electrode, paper separator, and iron-foil (−) electrode. For ongoing lighting and phone charging sessions, the user will purchase small pre-packaged quantities of consumable cell materials (carbon electrode, paper separator) and active materials (iron and supporting electrolyte salt powder, and iron or steel foil). The user dissolves active iron salt ($Fe_2(SO_4)_3$) and supporting electrolyte (NaCl) in locally-available water, and this solution is fed into the (+) electrode by gravity. The product will provide the user with clear instructions on how much water to mix with the dry chemical (for example fill lines on the product inlet tank or separate mixing cup). During discharge of the cell to produce power, the Fe^{3+} is reduced and the iron foil is oxidized, according to

$$Cathode: \quad 2Fe^{3+}(aq) + 2e^- \rightleftharpoons 2Fe^{2+}(aq) \quad E^0 = 0.77 \ V \ vs. \ SHE \tag{1}$$

$$Anode: \quad Fe^0(s) \rightleftharpoons Fe^{2+}(aq) + 2e^- \quad E^0 = -0.44V \ vs. \ SHE \tag{2}$$

$$Overall: \quad 2Fe^{3+}(aq) + Fe^0(s) \rightleftharpoons 3Fe^{2+}(aq) \quad E^0 = 1.21V \tag{3}$$

In the initial proof-of-concept work, the electrolyte solution species ($Fe_2(SO_4)_3$ and NaCl) were selected for low cost, high performance, solubility, and non-toxicity [6]. Porous carbon paper (Sigracet 10AA) showed the highest performance of several carbon papers tested, and was found to be a significant contributor to total system cost. Office paper was demonstrated to be an acceptable and very low-cost separator, and provides open circuit voltage (OCV) of about 1.0 V. Low carbon steel was selected as the (−) electrode iron source, as high-purity iron is not required. In the present work, we dramatically increase the cell performance in lab testing hardware by selecting improved paper separator and carbon porous electrode materials. One design goal is to minimize the size of the device by maximizing cell power. This helps to minimize up-front purchase price which for users in extreme poverty is a more difficult barrier than ongoing consumables cost. Our previous work indicates that power (and energy) are maximized for discharge at 0.55 V [6], so that is adopted as the standard test condition here. The cell materials selected in lab-hardware tests are then demonstrated in a proof-of-concept stand-alone consumer product prototype fabricated in plastic for low cost and designed for ease of use by the end-user.

2. Experimental methods

Electrolyte solutions (0.25 M iron sulfate and 1.2 M sodium chloride) were prepared by mixing de-ionized water with $Fe_2(SO_4)_3$ (Sigma Aldrich analytical grade) and NaCl (BDH analytical grade). Low-carbon steel (−) electrode sheets were provided by McMaster-Carr. Sheets were cleaned with acetone and polished lightly with sand paper before cell assembly to eliminate surface oxidation or machine oil contamination. Carbon porous electrode papers were provided by SGL (Sigracet 10AA) or CeraMaterials (all others listed in Table 1). Pre-treated carbon felt electrodes were soaked in concentrated sulfuric acid (Sigma Aldrich) for 1.5−20 h at room temperature. Treated felts were washed thoroughly with deionized water before cell assembly. Various paper separators were tested including office paper (printer and copier paper, Boise Cascade), laboratory filter paper (GE Whatman), and coffee filter paper (Melitta, If You Care, and generic).

Cells were assembled in standard fuel-cell-testing hardware (Fuel Cell Technologies) with 10 cm^2 active area. Flow-through Niobium flowfields were used on the positive side to avoid carbon oxidation or metal corrosion (Treadstone Technologies, Inc.). The carbon porous electrode and iron electrode were inserted in window-frame shape Teflon gaskets, the thickness of which was selected to compress the carbon porous electrode to the intended thickness, which varied between 35 and 85% of the original thickness. The iron thickness (0.15 mm) was selected to be significantly thicker than necessary, so that consumption of iron metal did not limit cell capacity. Electrolyte solution was provided by a syringe pump at 1 mL min-1 or by gravity feed (as indicated in the figure captions) and was not recirculated from the cell exit. The pump flowrate was chosen to mimic the flowrate typically observed with gravity feed.

Cells were tested at room temperature with a VMP-3 (BioLogic) potentiostat. Constant-voltage discharge at 0.55 V immediately followed cell assembly and solution introduction. Cell OCV was in the range 0.98−1.05 V in the several seconds before discharge commenced. Fe^{3+} utilization was determined from coulometry during cell operation. Efficiency was determined as the ratio of applied external current capacity (mAh) to total electrochemical activity (including applied current, self-discharge, corrosion, and any other side reactions that consume iron metal) assessed by weight loss of the iron foil.

Carbon felt re-usability was determined by testing and disassembling a cell, recovering the carbon felt, letting it dry 24 h in ambient conditions, and re-using it in another cell assembled with fresh paper, iron foil, and electrolyte solution. This was repeated for 7 total uses of the carbon felt.

A stand-alone prototype was designed with CAD software (SolidWorks), and fabricated using a 3D printer (Series 1 Pro, Type A Machines). A viton o-ring was used as a seal between the two halves of the screw-cap cell housing. The two cells were attached in series and connected to a printed circuit board (PCB) containing a white light-emitting diode (LED, Seoul Semiconductor STW8T16C) and LED-driver (Zetex ZXLD383).

3. Results and discussion

3.1. Carbon porous (+) electrode

Previous work with Sigracet 10AA carbon paper electrode demonstrated porous carbon to be an effective electrode for this system [6]. Here, a range of lower-cost highly porous carbon felts provided by CeraMaterials are examined. The felts are produced
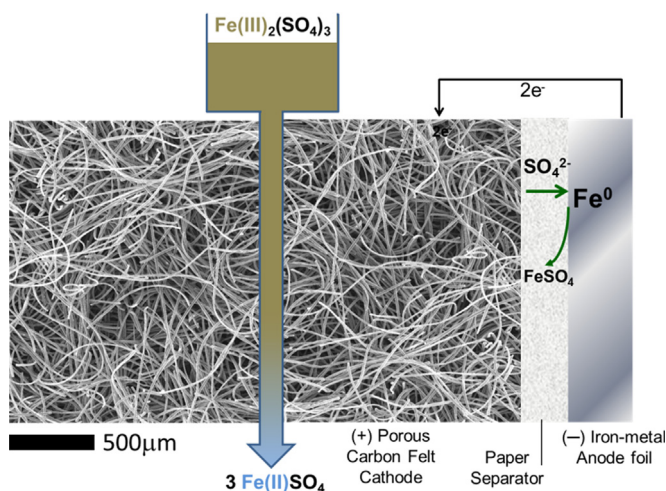

Fig. 1. Schematic of the all-iron flow cell. The horizontal axis in the image is to scale. The iron sulfate solution is introduced by gravity feed from an inlet reservoir, and flows through the entire thickness of the porous carbon cathode.

Table 1
Comparison of carbon paper (Sigracet 10AA) and various carbon felts.

	Thickness (mm)	Cost ($ m^{-2})	Avg. Power density (mW cm^{-2})	Power cost ($/W)
10AA	0.38	80	24	0.33
Rayon 1200 °C	3.2	38	26	0.15
Rayon 2000 °C	2.4	49	28	0.18
PAN 1200 °C	3.0	27	22	0.12
PAN 2000 °C	2.8	35	26	0.13

from either Rayon or polyacrylonitrile (PAN) fiber felts, and carbonized at 1200 °C or graphitized at 2000 °C. These felts have been demonstrated as effective electrodes for aqueous electrochemistry in a variety of systems including vanadium and zinc-bromine redox flow cells, microbial fuel cells, and hydrogen-ferric-ion recombination [7—10]. The structure of the felt is compared to Sigracet 10AA carbon paper in Fig. 2a. Whereas the carbon paper contains a high proportion of graphite particles, which fill many of the pores between the carbon fibers, the felt is a highly open structure (~95% porosity) consisting only of fibers. The felt is also about 3 mm thick, in contrast to the thin paper which is about 0.3 mm thick. Performance of the range of felts and carbon paper was determined by potentiostatic discharge of a Fe^{3+} solution, using a low flowrate chosen to reflect the flowrate expected from gravity feed in the final prototype device. Typical discharge curves are shown in Fig. 2b. A rapid initial transient occurs as the Fe^{3+} contained within the electrode is depleted, leading to lower stable current dominated by mass transport within the thick electrode, and a substantial state-of-charge gradient between the inlet and outlet of the (+) electrode. Note that the transient is much faster for the thinner carbon paper. Comparison of the various felts and carbon paper is shown in Table 1. All provided a similar steady-state current density in the range 22—28 mA cm^{-2}. The low cost of

the felts results, however, in significantly lower cost of power, a key metric for affordability of the proposed system. The Rayon 1200 °C felt was selected as the best because it provided among the lowest cost of power, and also was the easiest to use in terms of being flexible and producing minimal carbon powder during handling (which could clog tubes in the consumer product).

Porous electrodes are typically compressed to ensure good electrical contact between the electrode and current collector, and among fibers within the electrode. Over-compression, however, reduces pore volume available for solution flow, and can damage the electrode by fracturing carbon fibers [11]. As shown by the shaded region in Fig. 2c, there is a wide range of compression for carbon felt that provides power density close to the maximum achieved at 1.8 mm (55% of the original 3.2 mm thickness). This is in contrast to the carbon paper, for which there is a very narrow band where acceptable performance is achieved. This is an extremely important consideration for the envisioned consumer product, as compression will be influenced by the machining tolerances of the plastic housing parts, and how consistently the user assembles the cell materials from run to run (see Section 3.5 for discussion of the cell assembly by the end-user). Using felt, which is much more forgiving of variations in the absolute compressed thickness, allows for greater design flexibility and lower-cost cell housing fabrication than would be required when using carbon paper.

One drawback of the carbon felt is that it is not easily wetted by aqueous solutions in the as-received state, with a 135° contact angle to the felt surface. To overcome this, the simplest solution is to submerge the felt in water or electrolyte and mechanically wet the felt by compressing it several times by hand. While effective, this is not a user-friendly solution. Addition of low-cost surfactant (dish washing soap) to the electrolyte solution lowered surface tension sufficiently to enable gravity feed. Felts were also pre-treated by soaking in concentrated sulfuric acid followed by water washing, as this is known to improve wetting and performance
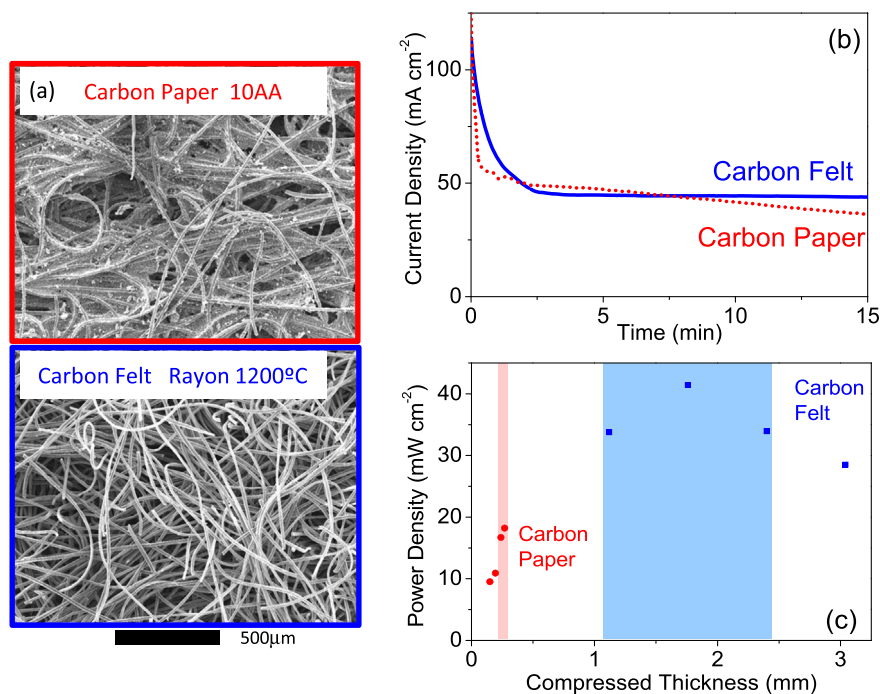
Fig. 2. Aspects of the porous carbon electrode. (a) SEM images of carbon paper (top) and carbon felt (bottom). (b) Potentiostatic discharge performance of Rayon 1200 °C carbon felt (solid blue line) and Sigracet 10AA carbon paper (dashed red line) at 0.55 V, 1 mL min^{-1} with laboratory filter paper separator. (c) Impact of compression on performance for Sigracet 10AA carbon paper and Rayon 1200 °C carbon felt. The shaded regions indicate the estimated range of acceptable compressed electrode thickness. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of carbon electrodes in other aqueous redox–flow-cell systems by introducing oxygen functional groups on the surface of the electrode [12,13]. This dramatically improved wettability, with solution quickly wetting through the entire felt when it was placed in contact with electrolyte solution. Fig. 3 shows performance of the cell with these various mechanisms for improving wetting. Soaking in sulfuric acid improved performance significantly, regardless of soak time, and was therefore chosen as the best approach for later experiments. Further reducing soaking time, and using diluted sulfuric acid are topics of future work.

Cost of the carbon paper (+) electrode was previously shown to be a large portion of the total system cost [6]. Therefore, reusing the (+) electrode is desirable to minimize operating costs. A carbon felt was reused daily, with all other consumables being replaced, to assess whether the felt is degraded by multiple cell assembly/disassembly cycles or multiple operation sessions. As shown in Fig. 4, no loss of performance occurred after a week of daily use. Note also that day-to-day performance variation is less than 9%. Longer-term studies and determining the impact of felt storage conditions (e.g. inside or outside the cell, wet or dry, etc.) will be the subject of future work.

3.2. Paper separator

Office (copier) paper was previously demonstrated to be an effective low-cost separator for this system, with cell performance inversely correlated to paper thickness due to ohmic impedance of ion transport through the paper [6]. To further reduce ohmic impedance, several types of filter paper were tested here. As shown in Fig. 5a, filter paper has a much more open pore structure, with large pores, compatible with its use in transporting liquid away from a bed of packed particles. Several types of coffee filter and one lab filter paper were tested, and the results are compared to office paper in Fig. 5b. Performance for both office papers increased during the first several minutes of operation, presumably due to slow wetting of the electrolyte into the dense paper. This did not appear to be an issue for any of the filter papers, as stable performance was achieved within 2 min after a typical initial decrease as discussed in Section 3.1. Coffee filter paper #3 provided the highest performance and was selected for subsequent experiments. It should be noted that all of the papers are weak and easily torn once wet, so we expect a fresh piece of paper will be consumed every
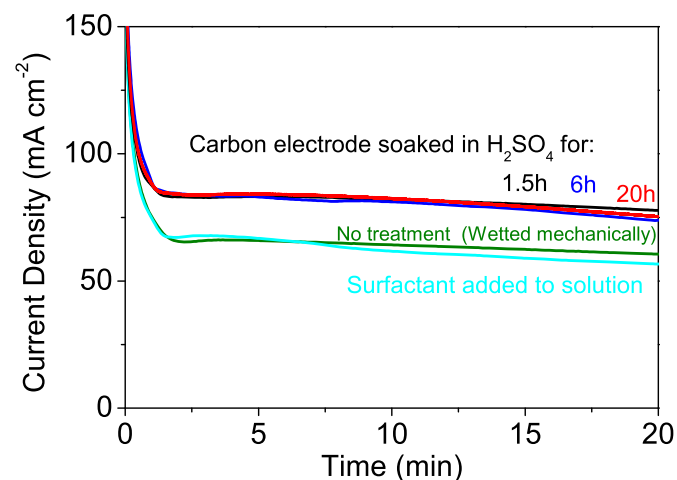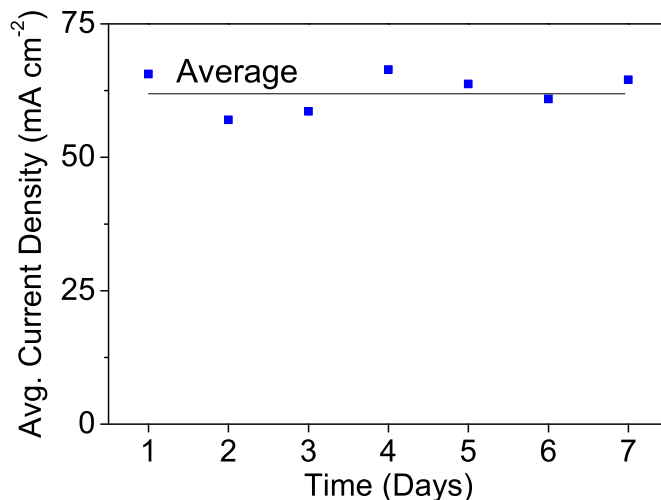

Fig. 4. Carbon felt durability. One week of daily re-use of Rayon 1200 °C carbon felt compressed to 75% of original thickness, 1 mL min^{-1}. Solution, filter paper, and iron steel foil were replaced each day.

time the cell is assembled for operation. The paper contributes minimally to the total consumables expense of the device, as discussed in Section 3.4, so this cost is easily accepted.

3.3. Full-discharge operation

The envisioned use case for this system is a single phone charge over approximately 2 h (2.7 W at 0.55 V) or a lighting session of 3–4 h (1.4–1.8 W at 0.55 V) for each set of consumable materials assembled into a cell. The experiments shown above were typically limited to 15 min, to capture the relevant short-term transient behavior and establish steady-state performance. To confirm that multi-hour operation is possible with this system, a cell was operated with a large single charge of electrolyte solution, as shown in Fig. 6. Continuous operation for 3.75 h was achieved, and the performance drops immediately once the inlet solution is depleted (around 225 min). The discharge profile is smooth, and no clogging of the flow path (for example by precipitates or carbon felt particles) is evident. The continuous decrease in performance is due to decreasing liquid column height, and therefore flowrate, as the electrolyte flows out of the feed tank into the cell. The bottom of the feed tank was approximately 5 cm above the cell inlet, and the liquid level was about 3 cm above this at the beginning of operation.

A cell was operated with gravity feed and the best paper (coffee filter #3) and best carbon felt (Rayon 1200 °C, H$_2$SO$_4$-treated) selected above. Volumetric energy density based on the volume of solution was 6.5 Wh L^{-1}. The performance is compared to the previous baseline in Fig. 7 and Table 2. Current density is dramatically increased due to the improved materials. The iron utilization is also improved. In the baseline case, electrolyte was continuously circulated through the cell by pumping at a relatively high rate (60 mL min^{-1}). This leads to nearly uniform state of charge in the (+) electrode, and low per-pass consumption of iron, resulting in relatively high Fe^{3+} concentration throughout the electrode. This leads to significant self-discharge via transport of Fe^{3+} through the paper separator to the (−) electrode [6]. Self-discharge consumes Fe^{3+}, lowering the achievable Fe^{3+} utilization. In contrast, the improved cell has a gravity-fed flowrate that is selected to nearly match the consumption of Fe^{3+} via the desired electrochemical reaction. Thus, the Fe^{3+} is nearly completely consumed in a single pass through the electrode, leading to low average Fe^{3+}

Fig. 3. Impact of various treatments to improve solution wetting into the electrode. Potentiostatic discharge performance of Rayon 1200 °C carbon felt at 0.55 V, gravity feed, compressed to 55% of original thickness, with coffee filter paper separator.
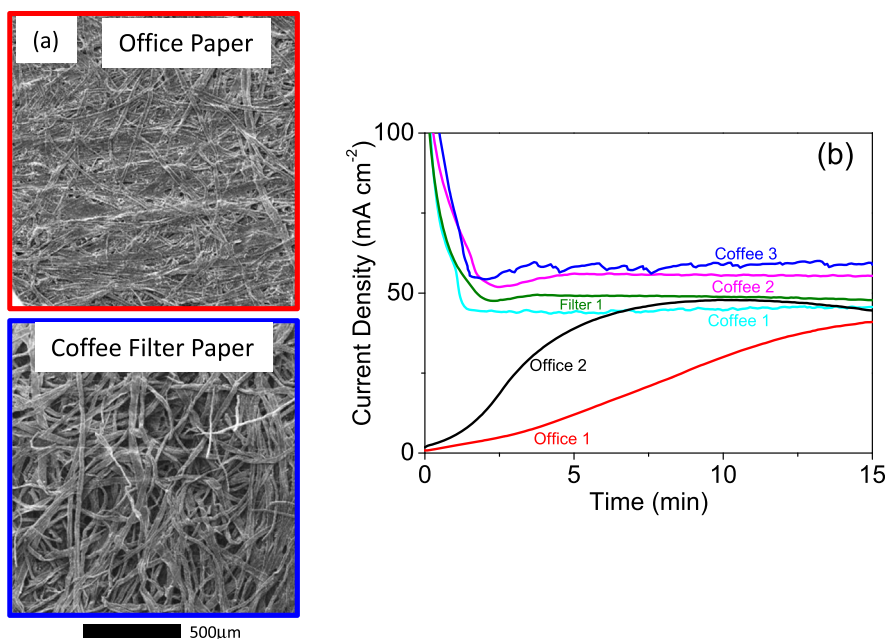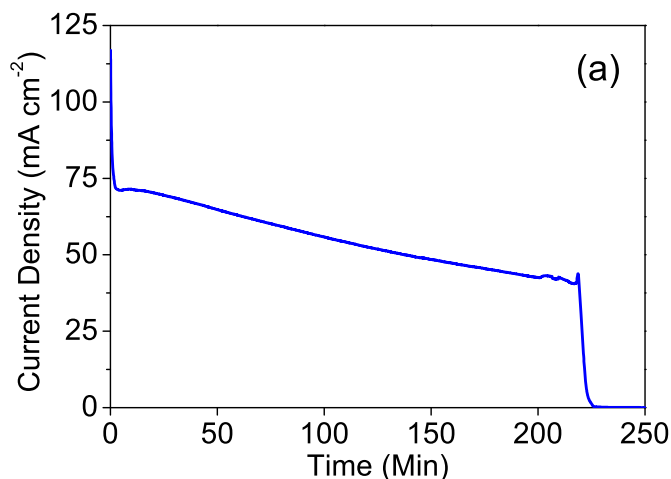
Fig. 5. Performance of various paper separators. (a) SEM images of office and coffee filter papers. (b) Potentiostatic discharge performance at 0.55 V, 1 mL min^{-1}, with Rayon 1200 $°C$ carbon felt compressed to 85% of original thickness.


Fig. 6. Use-case operation. (a) Long single potentiostatic discharge at 0.55 V with 200 mL solution, gravity feed flow, coffee filter paper and Rayon 2000 $°C$ carbon felt.

concentration in the electrode and therefore low self-discharge rate. For this situation, high coulombic efficiency is achieved, and utilization is limited by the residual amount of Fe^{3+} that exits the system.

3.4. Cost estimation

A use case for mobile phone charging was established and discussed previously for the baseline cell metrics in Table 2 [6]. That case is updated here to reflect the improved materials, performance and Fe^{3+} utilization. For a single phone charge, 5.4 Wh is required (2 h at 2.7 W). The consumables required to be purchased to deliver this energy are shown in Table 3. Additionally, the user must provide 0.83 L of water to prepare the solution; this is a reasonable volume compared to the many liters consumed daily for drinking and other household tasks. The total consumables cost is less than 3
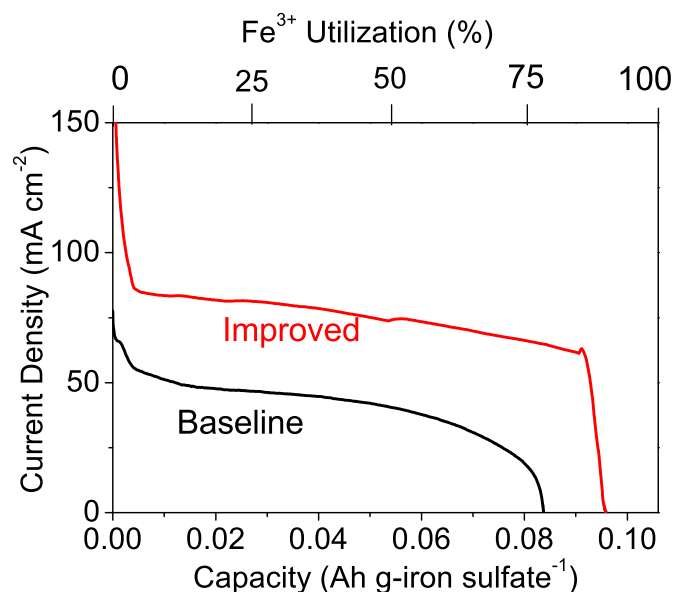
Fig. 7. Improved potentiostatic discharge performance at 0.55 V with gravity flow, Rayon 1200 $°C$ carbon felt pre-treated with H_2SO_4 for 6 h and compressed to 75% of original thickness, and coffee filter paper separator (#3). Baseline data from Ref. [6] is shown for comparison, with as-received Sigracet 10AA (+) electrode and office paper separator.

US cents, improved by about 15% from the previous baseline due to the increased energy density and Fe^{3+} utilization. This cost is dramatically lower than the $0.25 per charge paid at phone-charging kiosk businesses. The chemical costs are based on bulk purchase of industrial ferric sulfate as used in the water treatment industry ($250 per ton), and refined sodium chloride as used in the chemicals industry ($100 per ton). The impact, if any, of purity differing from the analytical-grade chemicals used in this study is a subject of future work. The weight and volume are also small and compatible with daily use. Cost savings is also achieved for the (+)

Table 2
Improvement in key operation metrics obtained from the data in Fig. 6, compared to baseline data from Ref. [6]. Energy density is based on weight of iron sulfate.

	Power density (mW cm^{-2})	Energy density (Wh kg^{-1})	Fe^{3+} utilization (%)	Efficiency (%)
Baseline	18	46	78	78
Improved	40	51	88	97

Table 3
Consumables required for a single mobile phone charge, assumed to be 5.4 Wh.

	Iron sulfate	Sodium chloride	Iron foil	Office Paper	Total
Weight (g)	84	12	12	0.9	**109**
Volume (cm^3)	26.9	5.7	2.0	1.5	**36**
Cost (US$)	0.021	0.001	0.006	0.001	**0.029**

electrode. By doubling the power density (Table 2) and halving the areal cost (Table 1), the (+) electrode cost drops from $1.20 to $0.26. This is a significant cost savings, as the durable plastic housing and electronic circuitry are expected to cost $2 to 3 at manufacturing scale. The electrode cost can further be spread over multiple energy production sessions as shown by re-using the carbon felt (Fig. 4). Furthermore, the required cell area is reduced by roughly half, leading to a more compact and lower-cost cell housing.
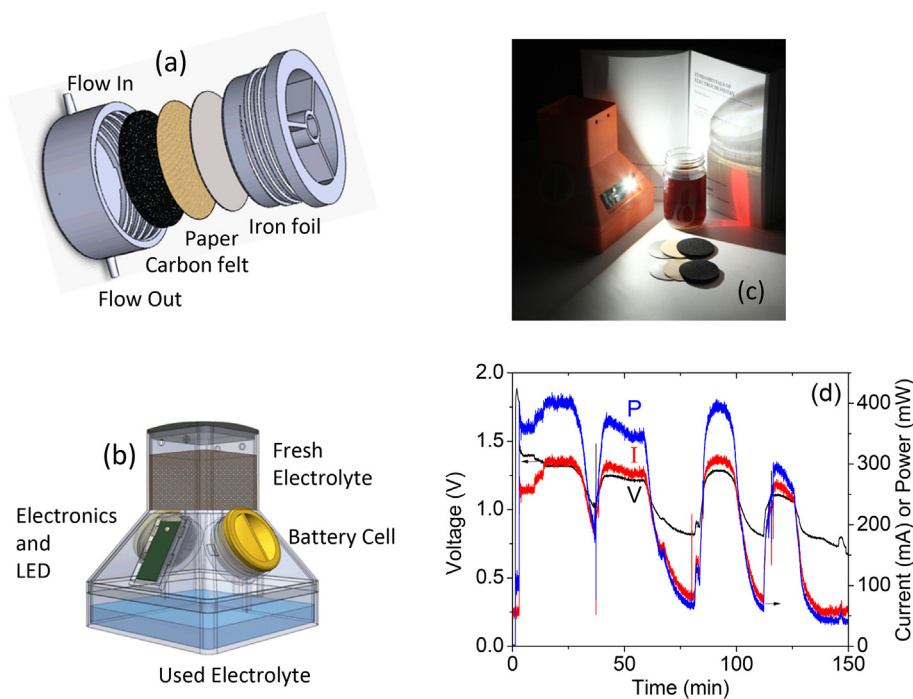
3.5. Consumer product prototype

Using the performance metrics and insights gained from the cells assembled in lab testing hardware above, we designed a free-standing proof-of-concept consumer prototype, shown in Fig. 8. For ease of assembly by end-users, a screw-cap cell housing was designed. The housing includes titanium contact foils as current collectors, which do not corrode in contact with the electrolyte solution. The cell materials are round to avoid the need for precise alignment (as would be required for square or rectangular components). The housing is slanted so that the cell materials do not fall out once they are placed in the housing. The cap is then threaded in until it bottoms out against an elastomer seal. The user clearly feels when the cap is fully inserted, ensuring a good seal as well as the correct compression of the carbon felt (although the compression is somewhat forgiving of variations in assembly, see Fig. 2c and related discussion).

Two cells are incorporated in the complete prototype (Fig. 8b). The cells are connected in series and provide power to an LED driver circuit with a single high-efficiency white LED of the same class used in inexpensive solar lanterns available in the developing world (Fig. 8c). After the cell materials are assembled into the screw-cap housings, fresh electrolyte is poured into the upper tank. It flows by gravity down through the cells, and into a lower holding tank in the base. The base is wide to prevent the unit from being knocked over easily.

The prototype was operated with two 20 cm^2 cells and 200 mL of electrolyte to produce LED lighting. The power provided by the cells (2 in series) to the LED driver is shown in Fig. 8c. The electrolyte flow was not well matched to the electrochemical consumption of Fe^{3+}, so the electrolyte had to be collected and returned to the upper tank four times to achieve complete discharge (indicated by color change of the solution from brown to faint blue). The goal is to have complete utilization in a single pass, to reduce user effort. This can be achieved by increasing the current (through optimization of the LED driver circuit), or reducing the flowrate (by altering the liquid column height or adding resistance to the flow path). It is also desirable to better match the cell area and LED driver characteristics so that the cells are operating close to the optimum 0.55 V. Despite the need for further refinement, this prototype clearly demonstrates the proof-of-concept, and provided

Fig. 8. Prototype consumer product proof-of-concept. Schematics of (a) screw-cap cell sub-assembly and (b) complete product. (c) Image of product in operation, illuminating a textbook and the consumable electrolyte solution and cell components. (d) Performance of two cells in electrical series, connected to a LED driver circuit. Solution (200 mL) was collected and returned to the top electrolyte reservoir 4 times to achieve complete discharge. Cells contained Rayon 1200 °C carbon felt pre-treated with H_2SO_4 for 1.5 h and compressed to 55% of original thickness, and coffee filter paper separator (#3).

2.5 h of modern light while using less than one cent worth of consumables.

4. Summary

A low-cost, non-hazardous all-iron electrochemical cell was improved significantly compared to previous work on this system. Improved performance was achieved by using coffee filter paper as the separator, and carbon felt treated by soaking in sulfuric acid as the (+) electrode. The pretreatment of the electrode also enables easy wetting of the electrolyte solution throughout the interior of the electrode. It was shown that the (+) electrode can be re-used many times to reduce operating cost. The cell improvements achieved here significantly reduce both the up-front and operating costs of the system, making it more accessible to the poorest consumers. A stand-alone proof-of-concept consumer product prototype was designed, fabricated, and tested. The prototype successfully provided a few hours of LED illumination using 200 mL of electrolyte solution. Future efforts should focus on optimizing the product design, and demonstrating long-term durability of the non-consumable components.

Acknowledgements

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