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Advancing Energy Storage: Sustainable Moldable Batteries with PVA and Rechargeable Copper Zinc Cells

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ABSTRACT

The increasing environmental toll of conventional battery production and disposal necessitates the exploration of sustainable alternatives. In this context, our battery emerges as an innovative solution, replacing rare and toxic metals with more abundant materials like copper and zinc. This shift in material utilization will markedly decrease the environmental degradation associated with the mining and processing of rare earth metals. With the imperative to curb environmental hazards from battery waste, hydrogel batteries stand out for their advantages such as moldability, efficient use of space, and reduction in material waste. Through analyzing the effects of various chemical and physical conditions our study produced a moldable PVA battery that can achieve both optimal voltage and charge retention to meet the standards of current batteries while harnessing many advantages for environmental safety and novel usage in modern batteries.

Introduction

Batteries are essential in modern life, powering various devices, from cell phones to electric vehicles. However, the failure to recycle batteries has far-reaching consequences for environmental and resource sustainability[1]. Due to the growing impact of pollution caused by mining of rare earth metals, there is a need to find other metal materials to create potent and strong batteries. The extraction and processing of rare earth metals such as lithium have long been associated with environmental pollution and degradation. Mining processes contribute to significant amounts of carbon emissions, with the production of lithium-ion batteries in China showing GHG emissions of 2,705 to 3,061 kg CO2-eq per 28 kWh battery, and the processes accounting for around 40% of total emissions being associated with electricity use[2]. The sustainability of these various mining industries is under severe concern. By utilizing abundant copper and zinc electrodes, our battery has the potential to be an eco-friendly alternative. Zinc-ion batteries are inherently safer and more recyclable due to the use of non-flammable aqueous electrolytes, making them a more sustainable choice overall[3].

Hydrogel batteries offer a number of advantages such as moldability, reduced weight, and superior elasticity and compressibility compared to solid-state batteries. These features are particularly needed for wearable devices, which require flexible, lightweight, and durable power sources to function effectively. The superior safety of hydrogel batteries is also a significant advantage, as they are less prone to leakage and thermal runaway, which are common issues in traditional batteries. Additionally, hydrogel batteries tend to be lower in cost, providing an economically viable option for various applications. They also possess adequate specific

[1] Cos et al., "Recycling and Environmental Issues of Lithium-Ion Batteries."
[2] Hao et al., "GHG Emissions from the Production of Lithium-Ion Batteries for Electric Vehicles in China."
[3] Zhang et al., "Fundamentals and Perspectives in Developing Zinc-Ion Battery Electrolytes."

capacity and other excellent electrochemical features, making them suitable for a wide range of applications beyond wearables, including medical devices and flexible electronics[4].

Notably, only a small fraction of non-lead batteries, less than 4 percent, are recycled[5]. This lack of recycling exacerbates the problem, as it's estimated that 51% of natural resources could be saved if effective recycling occurred, reducing fossil fuel and nuclear energy consumption in production[6]. But the environmental footprint of batteries extends beyond their end-of-life disposal. The extraction and processing of raw materials, including lithium, cobalt, and nickel, results in pollution and waste[7]. Furthermore, manufacturing generates greenhouse gas emissions[8]. The consequences profoundly impact the environment and human health[9].

In response to these challenges, we sought out to determine the most important factors to increase charge retention in moldable polyvinyl alcohol (PVA) hydrogel copper-zinc batteries. Our battery represents a significant advancement over traditional batteries, offering several key benefits that promote sustainability and environmental responsibility. This technology employs mechanical energy for recharging, reducing the environmental impact of battery production and disposal. By using a PVA and borax base, our battery creates a hydrogel that can adapt to irregularly shaped objects, thus saving space and materials. Varying concentrations of PVA and borax yield different consistencies of the hydrogel, enabling a wide range of applications. One unique advantage of PVA is its minimal contribution to microplastic pollution[10]. However, our battery does not rely solely on a PVA/Borax hydrogel; it can be used with any electrolyte that has a basic pH: NaOH for example. This allows for more versatility in the physical properties of the electrolyte, which can benefit the assembly and use cases for the battery. As a sustainable rechargeable battery, our battery addresses the pressing need to reduce the carbon footprint associated with traditional battery technologies, which generate waste and pollution[11].

Methods

Hydrogel Preparation

Borax and PVA were accurately weighed using a weigh boat and a scale within 2 to 10 grams, depending on the desired proportions. The Borax and PVA were then separately dissolved in 90-98 mL of a 6.25% NaCl solution. The solution's volume varied depending on each component's desired concentration. These solutions were vigorously agitated before further processing to ensure complete dissolution. Subsequently, the solutions were combined on a hot stir plate and stirred until a cloudy white gel, called the hydrogel, formed.

Hydrogel Storage

Any unused hydrogel was stored in airtight containers at ambient temperature (20°C) to prevent dehydration.

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Cell Setup and Voltage Measurements

To investigate the electrical properties of the hydrogel, we cast it into silicone molds to create four identical 25 mL cubes. Within each cube, a 5 cm copper strip and a 1.27cm zinc washer were connected in series. The cathode and anode terminals were then connected to a multimeter. The voltage measurements were recorded at several time intervals, including immediately upon assembly and after 1 minute, 5 minutes, 10 minutes, 15 minutes, 20 minutes, one day, two days, three days, and one week. Additionally, any abrupt voltage changes were carefully documented. The collected data was logged in a spreadsheet and subsequently utilized to generate graphical representations.

Recharging Station

For recharging the hydrogel-based batteries, a recharging station that employed four servos was designed, each with a stall torque of 17.2 kg at 4.8V, controlled through Arduino code. This recharging station was engineered to ensure simultaneous rotation of the four servos at a uniform rate. This synchronized motion allowed for equal pressure distribution to all four battery cells, facilitating recharging.

The provided code demonstrates how to control four servos (four cells) using Arduino:

```
#include <Servo.h>
Servo servo1, servo2, servo3, servo4;
int servo1Speed = 50;
int servo2Speed = 50;
int servo3Speed = 50;
int servo4Speed = 50;
void setup() {
  servo1.attach(9);
  servo2.attach(10);
  servo3.attach(11);
  servo4.attach(12);
}
void loop() {
  // Rotate servos in one direction
  servo1.writeMicroseconds(1500 + servo1Speed * 5);
  servo2.writeMicroseconds(1500 + servo2Speed * 5);
  servo3.writeMicroseconds(1500 + servo3Speed * 5);
  servo4.writeMicroseconds(1500 + servo4Speed * 5);
  delay(1000);
}
```

Utilizing the Servo library, this code facilitates the control of four servos. The setup() function initializes the servo connections, while the loop() function repetitively updates servo angles. The value 1500 represents the neutral position for continuous rotation mode. The code incorporates a 1-second delay between servo movements.

Results

Effect of PVA Concentration on Charge Retention and Maximum Voltage

In order to determine the effect of hydrogel composition on charge retention, we tested increasing concentrations of PVA in the hydrogel in order to increase the charge retention over a one week period. Charge retention was calculated by taking the average number of volts lost per minute over a 10,000 minute period. We observed increasing the concentration of PVA in the hydrogel batteries resulted in a lower charge retention across a range of PVA concentrations (Fig. 1A). For instance, the 10% PVA hydrogel resulted in about 0.00003 V lost/min, and 1% PVA resulting in the best charge retention of about 0.000009 V lost/min.We hypothesized that a higher PVA concentration would be required for maximum battery voltage; however, all conditions performed comparably with a non significant decrease seen in the 4% PVA condition. (Fig 1B). Therefore, we concluded PVA could be reduced down to 1% without significantly reducing the maximum voltage output in our hydrogel battery.

Determining The Optimal Properties for Testing and Designing Hydrogel Batteries

In order to provide the largest dynamic range to test recharging capacity, we tested multiple electrolyte solutions to identify which one would result in the most optimal maximum voltage. The 4% Borax solution similar to that used in our hydrogels only demonstrated a maximum voltage of 0.75V per cell. This max voltage was similar to that of a diluted Dawn dish soap solution and 5% Sodium Hypochlorite. The 1.25M 5% NaOH solution demonstrated nearly 30% higher maximum voltage compared to all other conditions tested. However, NaOH displayed the least efficient charge retention compared to the other electrolyte solutions, with the Diluted Dawn Dish Soap yielding the most optimal charge retention. Despite the poor charge retention of the NaOH electrolyte, it resulted in the highest maximum voltage and was chosen to further compare against the hydrogel batteries in hopes of optimizing recharge capability through analyzing how an electrolyte yielding a high maximum voltage compares to an electrolyte that results in an efficient charge retention. Therefore NaOH batteries were selected to next compare to moldable, flexible 4% PVA, 4% Borax, 6.25% NaCl batteries.

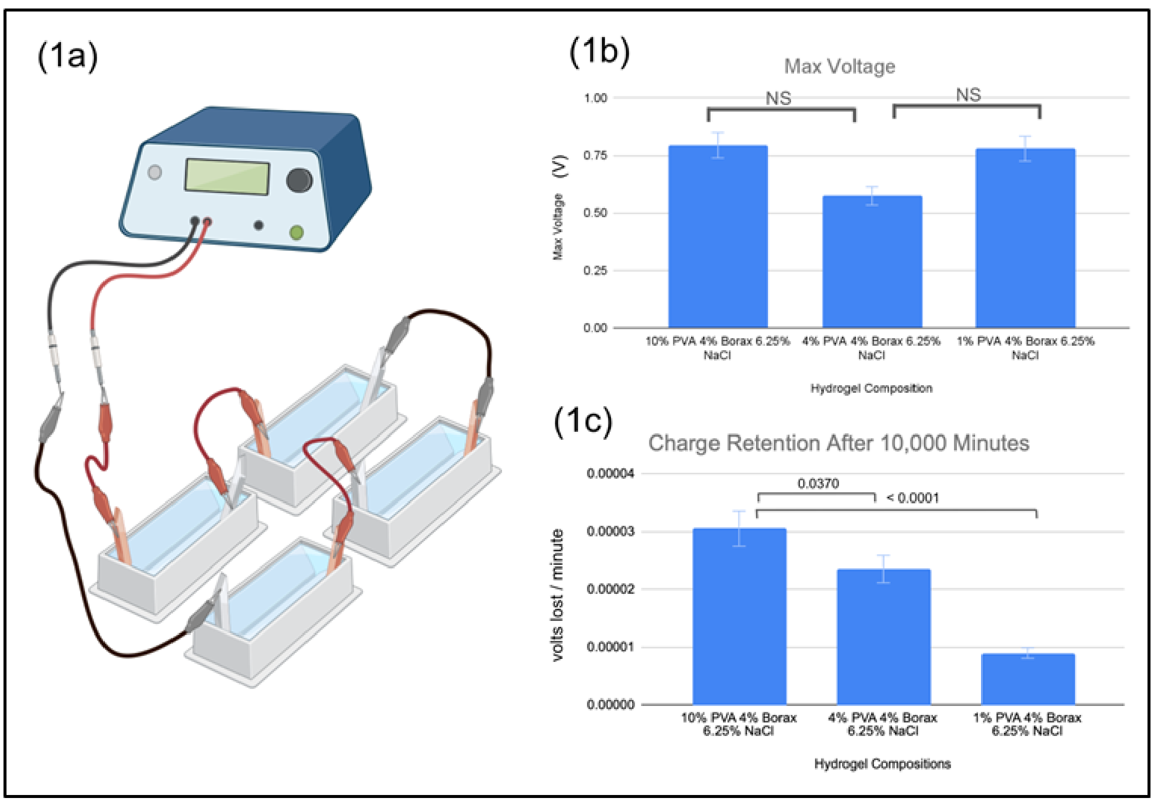

Figure 1. Voltage and Charge in Different Hydrogel Compositions.

(1a) Experimental setup of the four cell hydrogel battery in series connected to a voltmeter. A four cell configuration uses about 6 grams of copper and about 6 grams of zinc on average, each cell containing one strip of copper and zinc. The electrolyte is placed in between the electrodes. (2b) Various hydrogel compositions were tested. PVA% is indicated for each sample. PVA — 10%, 4%, and 1% — each with a constant 4% borax and 6.25% NaCl. The bar graph illustrates the maximum voltage (volts) achieved by each hydrogel battery. (3c) The bar chart compares the average voltage loss per minute of three different hydrogel compositions over an extended period of 10,000 minutes. The hydrogel compositions vary by the percentage of PVA used, each containing 4% borax and 6.25% NaCI. The compositions tested are 10% PVA, 4% PVA, and 1% PVA. Error bars on each bar denote the standard deviation from the mean.

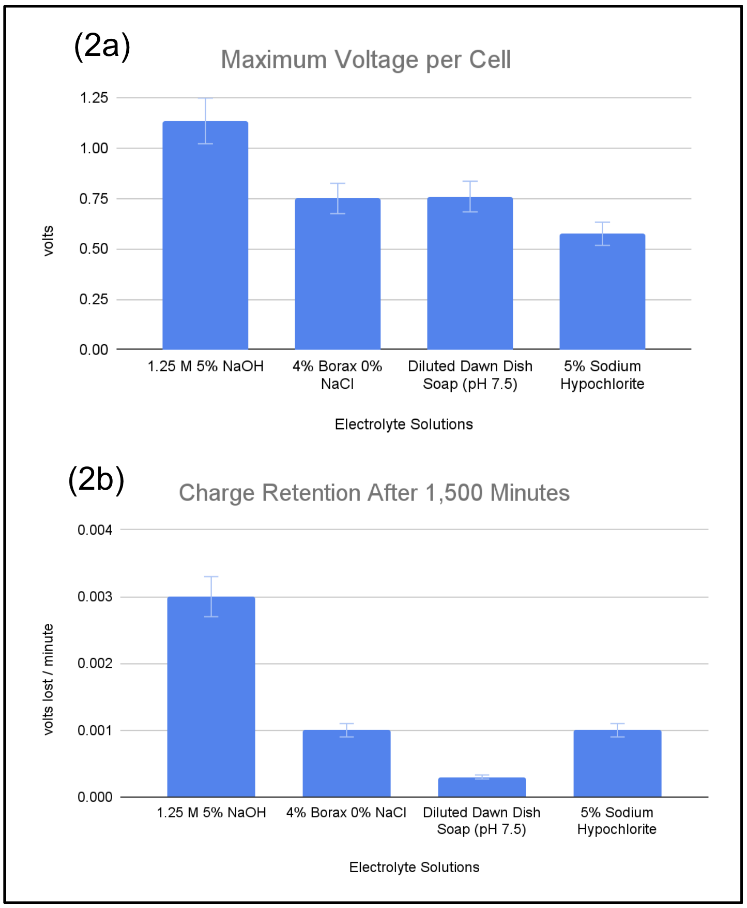

Figure 2. Comparing Maximum Voltage and Charge Retention in Different Electrolyte Solutions.

(2a) The figure depicts a bar chart showing the maximum voltage per cell for various electrolyte solutions. The solutions include 1.25 M 5% sodium hydroxide (NaOH), 4% borax with 0% sodium chloride (NaCl), a diluted solution of Dawn dish soap with a neutral pH of 7.5, and a 5% sodium hypochlorite solution. (2b) This bar graph illustrates the average voltage lost per minute for different electrolyte solutions over a period of 1,500 minutes. The electrolyte solutions tested include 1.25 M 5% NaOH, 4% Borax with 0% NaCl, Diluted Dawn

Dish Soap (pH 7.5), and 5% Sodium Hypochlorite. Error bars on each bar indicate the standard deviation, illustrating the variability in the maximum voltage readings across multiple trials or samples.

Our Hydrogel Batteries Demonstrated Superior Moldability While Performing Similarly to NaOH Batteries

In order to determine the recharging capabilities of our PVA battery, we set up a manual recharging station (Figure 3A) using a silicone ice tray to house the batteries. These experiments aim to showcase the difference in maximum voltage before and after recharging. We set up battery trays composed of either 4% PVA, 4% Borax, 6.25% NaCl (4% PVA hydrogel) or 1.25M NaOH. We measured the initial maximum voltage and the voltage post-recharge. Although the 4% PVA hydrogel had an initially lower maximum voltage compared to the 1.25M NaOH, the 4% PVA hydrogel had comparable recharge capability (Figure 3B). Additionally, the 4% PVA hydrogel recharged 39% of its initial voltage while the 1.25M NaOH recharged 49% of its initial voltage (Figure 3C). The NaOH can lead to environmental issues and increased corrosion due to its high pH. Since the recharge capacity of the 4% PVA hydrogel is comparable to the recharge capacity of the NaOH, a 4% PVA hydrogel electrolyte serves as a sustainable alternative to the NaOH, capable of replicating the optimal battery features. An automated recharge system offers several advantages. It would ensure consistency in recharging by eliminating the variability associated with manual processes. Such a system could deliver precise force and charge distribution to each cell, reducing the risk of overcharging or undercharging, which can degrade battery performance over time. Moreover, automation could potentially increase the number of charge cycles a battery can undergo while maintaining optimal performance, ultimately extending the battery's life and reliability in practical applications.

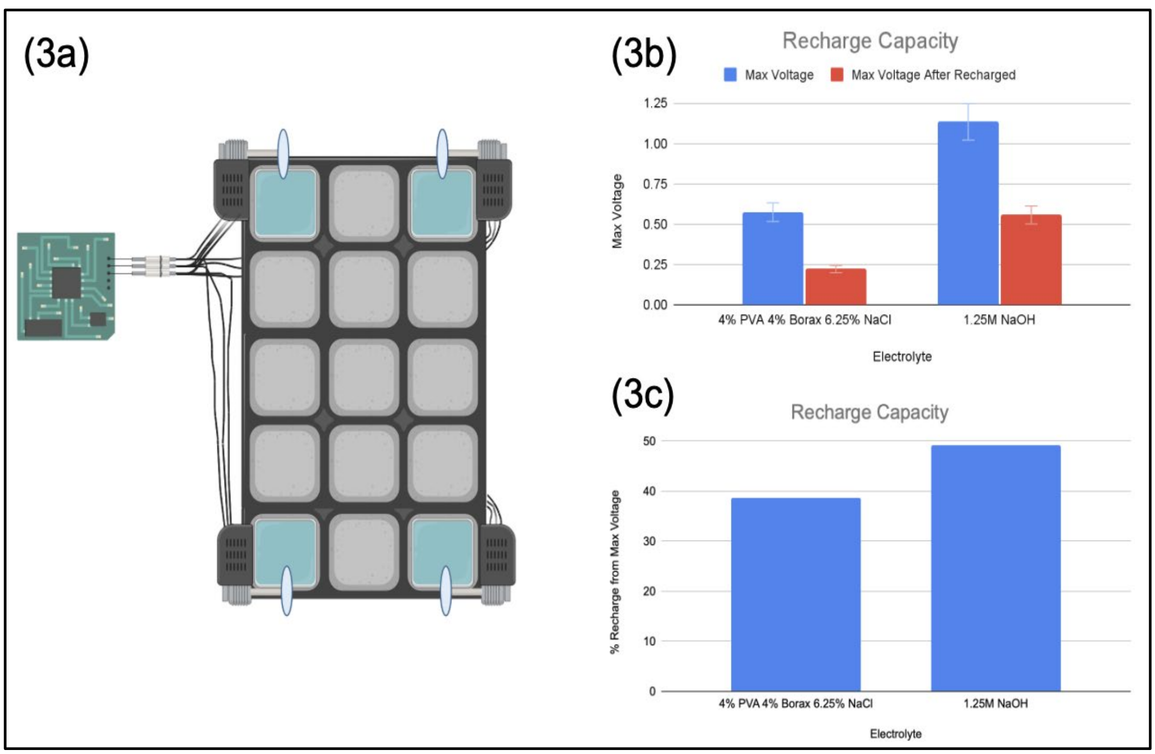

Figure 3. Comparing Recharge Capacity Between Hydrogel and Electrolyte Batteries.

(3a) Model setup for recharging hydrogel-based batteries, which employed four servos was designed, each with a stall torque of 17.2 kg at 4.8V, controlled through Arduino code. This recharging station was engineered to ensure simultaneous rotation of the four servos at a uniform rate. This synchronized motion allowed for equal pressure distribution to all four battery cells, facilitating recharging. (3b) This figure presents a comparison of recharge capacity between two electrolyte solutions: a 4% PVA 4% Borax 6.25% NaCl solution and a 1.25M NaOH solution. Two sets of data are shown for each electrolyte: the initial max voltage (blue bars) and the max voltage after the electrolyte has been recharged (red bars). The y-axis quantifies the max voltage in volts. The x-axis categorizes the bars by electrolyte type. Error bars indicate the standard deviation of the measurements, reflecting the variability of the max voltage readings. **(3c)** The bar graph illustrates the recharge capacity, expressed as a percentage of recharge from the maximum voltage compared to the maximum voltage after applying mechanical force, for a 4% PVA 4% borax 6.25% NaC hydrogel and a 1.25M 5% NaOH electrolyte solution.

Variability of Consistencies and Properties Across Different Batteries

The study investigated the impact of different hydrogel concentrations on their consistencies and physical properties, (summarized in Table 1). These variations also influenced the extent of corrosion, ranging from minor green "crust" formation to complete oxidation, where the metal was enveloped in green rust (Data Not Shown). Introducing NaCl to the hydrogel altered its properties, increasing density and reducing moldability. However, lower NaCl concentrations allowed for improved moldability. The composition of the hydrogel electrolyte has proven to work with diverse chemicals with basic pHs. In addition, a trial utilizing the incorporation of nanofibrillated cellulose displayed a more optimal shape-conforming capability. The 4% PVA hydrogel was much more dense than the 2% PVA hydrogel, allowing the 2% PVA hydrogel to retain its moldability after solidification, whereas the 4% struggled with moldability after the hydrogel solidified. The incorporation of NFC extended the amount of time it took for the hydrogel to solidify from about 10 minutes without NFC to one week with NFC. This led us to not include NFC in a majority of our studies. The investigation into different hydrogel concentrations has revealed significant insights into their consistencies, physical properties, and performance as battery electrolytes. These findings underscore the importance of carefully selecting hydrogel compositions to optimize battery performance and durability, with potential applications across various industries.

Table 1. Variability of Consistencies and Properties Across Different Hydrogel Concentrations

Electrolyte	Physical Properties & Observations
4% PVA 4% Borax 6.25% NaCl	• Hydrogel was moldable and flexible when first combined at a temperature of 100 degrees Celsius • As the hydrogel cooled down, it solidified and took the shape of the container that it was in • It was no longer flexible or moldable after it was completely solidified
4% PVA 4% Borax 0% NaCl (Distilled H_2O)	• After squeezing cells for an additional 5 minutes, the voltage was 1.80v, lighting up an LED • Without the NaCl, this hydrogel was much more flexible and moldable for a much longer period of time than hydrogels with NaCl • This hydrogel could've been sprayed using a spray bottle due to its viscous properties • However, the lack of NaCl resulted in reduced charge retention
10% PVA 4% Borax 6.25% NaCl	• This hydrogel was initially much more elastic and stretchy than 4% PVA and 2% PVA when hot, but after it solidified, it was very dense, unable to be molded
2% PVA 2% Borax 6.25% NaCl	• 2% PVA yielded half as much hydrogel • When the borax and PVA were first combined, the hydrogel was slightly flexible and moldable but not as much as the 4% PVA or 10% PVA • This hydrogel solidified more quickly
4% PVA 4% Borax 0% NaCl (Distilled H_2O) with NFC	• Very brittle • Over time it shrunk but stretched out to touch the walls of the container • It took about a week to fully solidify • Able to recharge
2% PVA 4% Borax 0% NaCl (Distilled H_2O) with NFC	• Not as brittle as 4% • Very gelatinous • Took about a week to solidify • Unable to recharge, but adding NaOH increased voltage immensely
1.25M 5% NaOH	• Had efficient charging speeds and capability • Poor charge retention • Able to recharge by shaking cell

Discussion

In order to demonstrate the power of sustainable battery technology with the potential to reduce environmental impacts substantially we have performed proof of concept testing in this study to reveal remarkable charge longevity, rechargeability, and adaptability in our novel PVA hydrogel batteries. The unique features of this battery, including the use of more abundant metals, its moldable properties, and its capacity to recharge through mechanical force, make it a compelling candidate for reducing the environmental footprint associated with traditional batteries. Nonetheless, challenges persist, primarily related to encapsulating the hydrogel within a hydrophobic capsule to ensure compatibility with electronic components. Our experiments revealed that the concentration of hydrogel has a direct impact on battery longevity and recharging time, with the 1% PVA 4% Borax battery resulting in the most optimal charge retention and the 1.25M NaOH battery resulting in the most optimal max voltage and recharge capability. We explored the idea of incorporating NaOH in our hydrogel battery with nanofibrillated cellulose (NFC) to optimize recharge capability, charge retention, and enhance moldable properties. The incorporation of NFC and NaOH resulted in a battery with enhanced shape conforming ability; however, we did not find any consistent improved voltage (Supplemental Figure 1A). Therefore we determined we were not going to include NaOH in the hydrogel because NaOH is highly basic and can lead to adverse effects such as: corrosion of electrodes and a toxic decomposition of the hydrogel. Furthermore, the concentration of NaCl and PVA affect the moldability of the hydrogel. By utilizing electrolytes containing low concentrations of NaCl and PVA, or by employing aqueous bases such as NaOH, the electrolyte can be sprayed over an area to reach the electrodes. This innovation could enhance battery assembly efficiency and enable creative product design, especially in devices with unconventional battery port dimensions. The moldable properties of our battery allow it to adapt to various object shapes and designs. Due to their lack of flexibility in design, traditional batteries often lead to inefficiencies in material use. The adaptability offered in our hydrogel battery can save space and reduce the need for additional housing materials, which can contribute to reduced environmental waste. The battery's moldable properties could revolutionize battery design, making it an excellent replacement for conventional batteries. Interestingly, one of our PVA batteries demonstrated an unusual pattern of behavior. Instead of experiencing a voltage drop due to the end of the chemical reaction, it was corrosion that led to the decrease in voltage because of a decrease in surface area. Replacement of the corroded copper and zinc components after a period of six days yielded a voltage that not only restored but actually surpassed the original voltage. This indicates that it was corrosion that primarily curtailed the battery's voltage capacity, not the depletion of the chemical reaction. Thus, curtailing corrosion could significantly prolong battery life and also enhance environmental safety by diminishing the dispersal of harmful substances.

Conclusion

The development of our PVA battery presents a groundbreaking opportunity to reduce the environmental impact associated with traditional batteries significantly. Its unique, moldable design conserves space and housing materials and introduces a sustainable alternative to conventional battery recharging mechanisms by harnessing mechanical force. Our results highlight the discovery of the most optimal PVA concentration in terms of charge retention and maximum voltage and its advantages over highly basic electrolytes like NaOH, underscoring the immense potential of our battery as a viable and sustainable substitute for conventional batteries. Our battery's exclusion of precious earth metals contributes to its ecological prowess, making it a cost-effective solution, offering a genuinely sustainable option to reduce our carbon footprint. Further research will focus on enhancing this technology by determining the tensile strength of each hydrogel concentration, resolving any corrosion concerns between the hydrogel and metal electrodes, developing a protective capsule to prevent gel dehydration, and changes in density.

Continued research and development of moldable, mechanically rechargeable batteries will lead to new and innovative applications. These advancements can address our pressing environmental challenges, marking a significant stride towards sustainable energy storage and comprehensive solutions. Our work opens up an exciting realm of possibilities in sustainable energy storage, undoubtedly contributing to the broader spectrum of sustainable energy solutions. The potential impact of this battery, both in terms of environmental conservation and technological innovation, is boundless.

Limitations

We understand that significant limitations are present when considering corrosion in affecting battery behavior, therefore we attempted control for this wherever possible and we performed a detailed analysis of corrosion during each of our experiments. Further research will be important to understand the potential impact of corrosion and limitations it poses when using novel hydrogel batteries

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