A novel piezoelectric sensor for continuous monitoring of sodium concentrations in sweat

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Abstract

Hyponatremia in athletes contributes to the onset of severe cardiac and neurological disorders; therefore, the continuous monitoring of sodium concentrations in the body is essential. Current devices utilize pilocarpine iontophoresis to enhance perspiration, which is limited to a usage time of five minutes and results in severe skin burns. This study proposed a piezoelectric cantilever beam sensor for continuous sodium concentration monitoring in human perspiration under various pathological conditions. The operating principle of the proposed sensor is that alterations in sodium concentration results in electric potential variation, which produces a measurable acceleration at the free end of the beam sensor. The effects of temperature, sweat rate, and volume on the sensitivity and efficacy of the sensor were investigated. Sodium concentrations ranging from 10 mmol/L to 130 mmol/L resulted in a linear decrease in voltage from 1.8 V to 0.31 V, which decreased acceleration from 149.74 mm/s$^2$ to 14.99 mm/s$^2$. Acceleration was found to have a sinusoidal signal with a frequency of 20 Hz, indicating that the sensor efficacy was not influenced by environmental interference frequencies. An increase in temperature from 22°C to 40°C resulted in a linear decrease in electric potential from 2.04 V to 0.8 V, indicating that a temperature sensor must be integrated into the sensor to prevent inaccurate measurements. Electric potential was constant as sweat rate and volume increased, demonstrating that the piezoelectric sensor surpasses past developments. Design dimensions can be reduced by integrating a MEMS accelerometer for enhanced employment by athletes.
**Personal Section**

One weekend, I was out for a game of soccer with my friends. The temperature was a blazing 95°F, and I began to feel light headed. I thought to myself: “Why am I experiencing these symptoms?” During halftime, I checked Google on my phone, and I learned that as I played, I excreted fundamental electrolytes, such as sodium, and water. However, I was only ingesting water, which resulted in a decrease in my body’s sodium levels, also known as hyponatremia. This created an osmotic imbalance across my blood brain barrier; therefore, a water flux caused my headache. After further research, I learned that my dizziness was just the tip of the iceberg—sodium deficiencies result in a host of cardiac and neurological complications. I also saw a headline from a soccer player at Ajax, Abdelhak Nouri, who had the same deficiencies as me, but his were far more serious—a cardiac arrhythmia led to permanent brain damage. This was when I realized that it was essential to develop a method for continuously monitoring sodium in athletes. I read countless journals to gain an understanding of current sensors, and I realized that their applications could not be realized due to drawbacks such as inaccuracy under low sweat rates and sweat volumes. From my knowledge of piezoelectric materials, I recalled that the energy outputs tended to have very small currents—currents small enough to not be felt by a person. Therefore, I came up with the concept of applying the small currents generated by the conductivity of sodium to deform a piezoelectric beam and evaluate if this sensor could address the limitations of past sensors.

My initial interest in piezoelectric materials began during my sophomore year. I was practicing my jazz solo during a soundcheck, and my director told me, “Ishan, aim towards the microphone. If you’re bell is in the stand you’ll end up harmonizing yourself.” While he was clearly joking, I began thinking how cool it would be if I could develop a way to play multiple notes simultaneously. When I searched for such a method on Google, I stumbled upon piezoelectric materials. Something inside of me clicked, and I knew that I wanted to dedicate my high school research career to advancing the field. I contacted Professor James West to request a placement in his lab. I am grateful that he met me when he was at a conference to discuss why piezoelectric materials are limited in large scale energy harvesting (miniscule currents reduce their power outputs), and all of the potential applications of the technology, which further spurred my interest.

The experimental stages of my project were performed at the Energy Harvesting and Mechatronics Research Laboratory at Virginia Tech under the direction of Professor Lei Zuo. However, I studied piezoelectric materials far before arriving. I had found material science textbooks online, along with chapters on piezoelectric materials, where I learned about constitutive equations and the differential equations that can be used to derive them. Understanding just the basic concepts of this required me to independently study the individual piezoelectric constants, their respective vectors, and how matrix algebra can be used to simplify them. Furthermore, I delved into medicine and biology to help understand
the physiological aspects of my project by self-studying medical textbooks my mother provided me with. While difficult and requiring extensive time to truly understand concepts, learning such advanced physics has heightened both my understanding and fascination of mathematics and the natural sciences. Further, realizing their intersections and cross-applicability has been truly rewarding and inspiring.

For high school students looking to undertake a research project, the one thing that is imperative to understanding is that if it were easy to do something, then everyone would do it. However, by pushing yourself past the initially daunting and difficult tasks, you will realize that all the blood, sweat, and tears are worth it as you will have realized a tool or concept that is invaluable to our understanding of the world around us, and to the development of our society. It’s not an easy road, but you need to continue to believe in your knowledge and potential, because if you don’t, then there is no reason for anybody else to.
Introduction

Hyponatremia (serum Na⁺<135 mEq/L) is evident in 30% of Ironman Triathlon competitors. With this statistic extrapolated to the endurance athlete population, millions of athletes face the risk of hyponatremia every year [1], [2]. Due to sodium’s instrumental role in the Renin-angiotensin-aldosterone system (RAAS) and the Na⁺/K⁺ ATPase pump, hyponatremia can disrupt many homeostatic functions [3]–[6]. Heart functions are also impeded since sodium imbalances interfere with myocardial action potential, which causes arrhythmias [7], [8]. Furthermore, hyponatremia changes the water balance across the blood brain barrier. This leads to a rapid influx of water into the brain, which can result in Traumatic Brain Injury (TBI) [9]. Such afflictions can cause irreversible damage to an athlete, which is why it is essential to develop a method for continuously monitoring sodium concentrations.

Current Methods

The conventional methods for sodium concentration monitoring are electrolyte pH panels and urine tests [10]. An electrolyte sweat test is primarily used for cystic fibrosis diagnosis [11], [12]. Sweat rates are increased by applying a current across the skin to expedite the skin’s absorption rate of the drug pilocarpine, increasing sweat diffusion for five minutes. Sweat is then harvested for thirty minutes and delivered to a hospital for examination. However, these existing methods fail to provide real time analysis of sodium sweat content since only a single fluid sample can be collected. A trained technician must then perform extensive laboratory analysis using cumbersome equipment. Primarily, pilocarpine during an electrolyte sweat test is linked to severe reddening and burning of the user’s skin. Due to these limitations, these methods cannot be used in athletes, who experience constantly fluctuating sweat sodium levels in a dynamic environment.

Therefore, noninvasive sensing techniques that analyze the sweat sodium content have been investigated [13], [14]. Recently developed wearable sensor arrays are advantageous due to their ability to monitor multiple analytes [15]–[17]. Such sensing arrays were fabricated using a plastic based sensor and silicon circuits to enhance flexibility while maintaining durability and are developed with the intent of monitoring glucose, sodium, and potassium levels in sweat. While this technique is advantageous due to its ability to accurately analyze various substances present in perspiration, it was primarily evaluated under vigorous athletic conditions, exhibited poor efficiency under low concentrations and low sweat rates, and had decreased accuracy after four uses, which mandates costly replacement [18]. Furthermore, these studies required pilocarpine to supplement sweat induced by exercise, which leads to burning of the skin [19]. Therefore, it is imperative that another method is devised.

Piezoelectric Sensors

Piezoelectric materials have recently demonstrated capabilities for energy harvesting from body motions and sensing of respiration and heart rates. Notably, the contraction and relaxation of the heart,
lungs, and diaphragm were harvested by a piezoelectric device to provide a renewable power source for pacemakers [20]. Furthermore, piezoelectric sensors have been used as an alternative ECG monitor, because changes in material deformation influence the resulting voltage output [21], [22].

These applications utilize the direct piezoelectric effect—mechanical deformations align the dipole moment of a piezoelectric material, resulting in a repolarization of the material and the production of a voltage of the opposite polarity (Fig. 1). Due to the electrolytic conductivity of sodium, a voltage applied across the solution generates a potential difference. This makes it possible to take advantage of the inverse piezoelectric effect (the material mechanically responds to an input voltage) because deformations allow variations in the potential difference to be measured, yielding a mechanism to detect changes in sodium concentration [23].

Among available piezoelectric materials, Lead Zirconate Titanate (PZT) has been utilized most frequently due to its high-power output and power generating capabilities. The inverse piezoelectric effect is evident in PZT because its crystal structure lacks a center of symmetry. Recent studies have utilized Polyvinylidene fluoride (PVDF) because along with being organic, biocompatible, and inexpensive, PVDF has a very high voltage coefficient [24], [25]. However, PVDF has a higher sensitivity than PZT, which is supported by its ability to be used in low energy applications [26], [27]. Both materials have been extensively evaluated in energy harvesting and sensing applications, but their properties have never been elucidated in response to electrolytic conductivity.

Therefore, the purpose of this study was to design and fabricate a piezoelectric sweat sensor to measure changes in potential difference caused by the variation of sodium concentrations and diagnose sodium imbalances. This study proposes, for the first time, the novel idea of deforming a piezoelectric beam from voltage induced by conductivity of sodium in sweat. The PZT sensor and a PVDF sensor are compared in this paper to confirm which material is more viable for this application. Lastly, the efficacy of PZT under varying sweat concentrations, volumes, and temperatures is evaluated in order to address past limitations.
Methods

Sweat Absorption Pad Fabrication

A 4 cm x 4 cm gauze pad was adapted to fabricate the sweat absorption pad. Two copper wires were secured to the pad at a height of 2 cm using a 1 cm x 0.5 cm strip of copper tape and separated by a distance of 5 mm. Adhesive tape was used for added reinforcement (Fig. 2).

The sweat pad connected in series to a 51 kΩ resistor to ensure that the current does not harm users. A sinusoidal voltage of 10 V (f=20 Hz) was applied using a Keithly 3390 50 MHz Arbitrary Waveform Generator. A Tektronix TDS 2014C Oscilloscope was connected in parallel to the sweat pad to measure the potential difference induced by sodium (Fig. 3a-b).

One of the key components of this experimental setup is realizing that the piezoelectric beam takes the place of a second resistor. While it could be argued that a potentiometer could just be used with a standard resistor, it must be noted that the current is in the micro and nano scale. Therefore, more cost effective and accurate measurements can be taken with a piezoelectric beam, which typically generates nano currents.

Figure 3. Sweat analysis setup. A) The sweat pad was connected in series to a 51 kΩ resistor and in parallel to an oscilloscope in order to measure the potential difference. B) Simplified diagram highlights interaction between components of the setup.

Acceleration Measurements

A Mide QP16n PZT cantilever beam was connected in parallel to the sweat pad, and the laser of a Polytech PSV 500 scanning vibrometer was directed towards the free end of the beam to measure acceleration. A low bypass frequency (f=40 Hz) was implemented in order to mitigate the effects of
ambient noise and natural air convection, which can be factors in sensor accuracy. PVDF (Tisch Scientific), was fabricated into a 3 cm x 1 cm beam. The PVDF beam was connected in parallel to the sweat pad and evaluated using the same methods as PZT.

**Synthetic Sweat Production**

The sweat solution was created using 0.1 micron filtered distilled water (Thermo Fisher Scientific). Sodium Chloride (Fisher Chemical) quantities ranging from 0.27 g to 3.79 g were dissolved into 500 mL of distilled water, yielding sweat solutions ranging from 10 mmol/L to 130 mmol/L Na\(^+\) that mimicked a wide range of pathologically relevant concentrations. The sweat solution was maintained at 37°C and a Thermix Stirring Hot Plate Model 210T maintained homogeneity of the sweat solution.

**Sweat Sensor Efficacy Evaluation**

**I. Concentration Analysis**

A standard dropper was used to deposit 10 mL of the sweat solution onto the sweat pad. An initial concentration of 10 mmol/L Na\(^+\) increased by intervals of 10 mmol/L until 130 mmol/L Na\(^+\). Variation in the potential difference was measured using the Tektronix Oscilloscope. Repeated in triplicate.

**II. Temperature Analysis**

Temperature of the sweat pad was measured using an Agilent U1242B True RMS multimeter. A dropper was used to deposit 10 mL of the sweat solution (50 mmol/L Na\(^+\)) onto the sensor. The temperature ranged from 22°C - 40°C in order to evaluate a wide range of pathological conditions. The potential difference was measured using a Tektronix Oscilloscope. Repeated in triplicate.

**III. Sweat Rate Analysis**

Initial sweat rate was 25 mL/hr and increased by intervals of 25 mL/hr until 1 L/hr. Different sweat rates tested for solutions of 30 mmol/L, 50 mmol/L, 70 mmol/L Na\(^+\), since 30 mmol/L Na\(^+\) indicates hyponatremia, 50 mmol/L Na\(^+\) is average concentration, and 70 mmol/L Na\(^+\) indicates hypernatremia. The voltage difference was measured using the Tektronix Oscilloscope. Repeated in triplicate.

**IV. Volume Analysis**

The 50 mmol/L Na\(^+\) was deposited on the sensor at a rate of 100 mL/hr. The potential difference measured using the Tektronix Oscilloscope was recorded as a function of volume at intervals of 0.5 L. Repeated in triplicate.

**Results/Discussion**

**Concentration Analysis**

The relationship between Na\(^+\) concentration and potential difference was evaluated to provide a voltage standard. A sinusoidal voltage (10V, f=20 Hz) was applied to a concentration gradient ranging
from 10 to 130 mmol/L. The initial concentration of 10 mmol/L resulted in a potential difference of 1.8V which decreased linearly until the final concentration of 130 mmol/L yielded a potential difference of 0.31V (Fig. 4).

Figure 4. Effect of Concentration on Potential Difference of Piezoelectric Sweat Sensor. Potential difference was found to decrease linearly as concentration increased from 10 mmol/L Na\(^+\) to 130 mmol/L Na\(^+\). Potential difference found using an oscilloscope, and all measurements taken at 37°C. Linear decrease indicates concentration can be measured using piezoelectric sensor.

The voltage standard produced provides a basis for comparison so that when future acceleration outputs indicate voltage, sodium concentrations can be found, allowing for accurate sodium monitoring. Greater sodium concentrations increase sweat conductivity, resulting in a decrease in resistance and potential difference. Furthermore, a distinct potential difference for each concentration indicated that a piezoelectric sweat sensor can be used to accurately detect changes in sodium concentrations.

**Acceleration Analysis**

The acceleration of a PZT beam was evaluated as a function of potential difference in order to provide a conversion for concentration measurements. The PZT was connected in parallel to the sweat pad and the laser vibrometer was focused on the beam free end. As the voltage increased from 0.31 V to 1.8 V, there was acceleration increased linearly from 14.99 mm/s\(^2\) to 149.74 mm/s\(^2\) (Fig. 5a). Using the voltage standard, a linear decrease in acceleration corresponded with an increase in sodium concentration from 10 mmol/L to 130 mmol/L (Fig. 5b). The acceleration for the 30 mmol/L Na\(^+\) sweat solution was found to be 126.16 mm/s\(^2\) and demonstrated a sinusoidal waveform (f=20 Hz) over a one second interval (Fig. 5c). When the 50 mmol/L Na\(^+\) sweat solution was evaluated, the resulting accelerations decreased to 90.79 mm/s\(^2\) and demonstrated a sinusoidal waveform (f=20 Hz; Fig. 5d).
The acceleration analysis showcases the operating principle of the sweat sensor, and also demonstrates, for the first time, that the potential difference induced by electrolytic conductivity of sweat can displace a piezoelectric cantilever beam. The acceleration decreased as concentration increased due to the decrease in applied potential difference, which ultimately decreases the magnitude of beam deformation. These accelerations can provide a basis for comparison so that the acceleration can be used to accurately measure sodium levels in athletes. Furthermore, the waveforms were sinusoidal due to the applied alternating current and had a frequency of 20 Hz because that was specified by the signal generator. Since the vibrometer was tuned to a low bypass frequency, the effects of destructive environmental frequencies (convection and sound) did not excite the beam. While PZT has been opposed due to lead content, these results demonstrate that PZT has strong capabilities for biomedical sensing applications.
**PVDF Evaluation**

Another potential material is PVDF, because PVDF has previously been utilized in biomedical applications, primarily as a pressure sensor for motions of the heart [28]–[30]. In this evaluation, the inverse piezoelectric effect was induced on PVDF when a potential difference was applied by a 50 mmol/L Na⁺ sweat solution. The resulting acceleration was shown to be inconsistent, random, and lacking a sinusoidal waveform signal, which indicates that the potential difference induced by sodium was not the primary cause of acceleration (Fig. 6).

![Graph showing acceleration over time](image)

**Figure 6. Drawbacks of PVDF actuator.** A 3 cm x 1 cm PVDF actuator was found to have poor sensor properties, as shown by the inconsistent waveform. The lack of a stable signal affirms that PVDF cannot sustain ambient vibrations with low bypass frequency, indicating that PZT is a more viable material.

PVDF is widely regarded as an extremely flexible material [31]. For that reason, it has also demonstrated high sensitivity when used for wind, rain, and acoustic sensing [16], [26]. Since PVDF has high sensitivity for environmental factors, it can be deduced that natural air convection and ambient noise, among other potential factors, increased the acceleration of the beam. This indicates that PVDF cannot differentiate between frequencies despite a low bypass frequency being used. Therefore, PZT is a more viable sensor than PVDF for this application.

**Temperature Analysis**

Temperature has shown to affect the readings of past sensors due to increased electrolyte conductivity. The temperature was set between 22°C to 40°C, in order to simulate a range of natural skin conditions. As temperature increased from 22°C to 40°C, the potential difference decreased linearly from 2.04V to 0.8V (Fig. 7).
The relationship between temperature and potential difference supports past studies that concluded temperature is a significant factor for sweat sensor readings [15]. Since temperature is a key factor in conductivity, due to enhanced kinetic energy, an increase in temperature increased the speed of the molecules. Due to their inverse relationship, an increase in conductivity reduced resistance, which caused a decrease in potential difference. While temperature is still a factor in sweat monitoring when a piezoelectric sensor is used, understanding the relationship between temperature and potential difference is essential for future investigations. The relationship demonstrates that a temperature sensor should be integrated into the sweat sensor design in order to calibrate the sensor response and prevent inaccurate diagnoses of sodium imbalances [25].

**Sweat Rate Analysis**

The next evaluation of the sweat sensor aimed to assess the feasibility of the sensor under varying sweat rates in elucidate the necessity for pilocarpine in a piezoelectric sensor. Additionally, this test aimed to demonstrate the abilities of a piezoelectric sweat sensor to work when sweat rates mimicked that of a sedentary person and an athlete. Sweat rates were evaluated for three 30 mmol/L, 50 mmol/L, and 70 mmol/L Na⁺. Each concentration demonstrated similar trends: there was a minor potential difference fluctuation (μ=0.08V) when sweat rate was below 100 mL/hr, but potential difference remained relatively stagnant until sweat rates equaled 1000 mL/hr (Fig. 8a-c).
The piezoelectric sweat sensor can work under low sweat rates. As compared to flexible sensor arrays, pilocarpine and vigorous athletic activity are not required. The potential difference remained constant with changing sweat rates because the Thermix stirring hotplate maintained the homogeneous solution. Further, pilocarpine iontophoresis induces sweat rates averaging 126 mL/hr, while this sensor was functional with a sweat rate of 25 mL/hr, demonstrating that this sensor is more efficient, does not pose a threat to users, and has a wider range of applications [32].

**Volume Analysis**

Sensor efficiency under increasing volumes was evaluated. The 50 mmol/L Na⁺ solution was applied at a sweat rate of 100 mL/hr, since that is when potential difference stabilized. Volume analysis began at 0.5 L and increased by 0.5 L until 12 L. Throughout the entire test, the potential difference was found to remain relatively constant as volume increased (Fig. 9).

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**Figure 8. Relationship between sweat rate and potential difference of sweat sensor.** a) Sweat Concentration of 30 mmol Na⁺. b) Sweat Concentration of 50 mmol Na⁺. c) Sweat Concentration of 70 mmol Na⁺. The potential difference was found to remain constant, with minor fluctuations when sweat rates were below 100 mL/hr for all concentrations. All figures indicate that the device can be used without sweat inducing mechanisms.

**Figure 9. Effect of sweat volume on potential difference.** Potential difference was found to remain relatively stagnant as the volume of sweat the sensor was exposed to increased. This indicates not only that the sweat sensor can function with low volumes, unlike past studies, but can work for extended periods of time.
The sensor is able to operate without induced sweat and has an extensive lifespan. If applied to an athlete, who can perspire up to 1 L per hour under most vigorous athletic conditions, this sensor can operate efficiently under 12 L of perspiration, indicating a lifespan of up to 12 hours. This is three times the lifespan of a flexible sensor array, which must be replaced after four hours of usage under similar conditions. Additionally, the sensing abilities of a piezoelectric sensor were further defined, and indicates that the amount of sweat does not impact the potential difference output [33]. Therefore, this confirms that the proposed sensor can provide a cost effective and safe method for continuous sweat monitoring.

**Conclusions**

This study investigated the novel application of the inverse piezoelectric effect by elucidating the response of a piezoelectric cantilever beam resulting from the electrolytic conductivity of sweat. By taking advantage of the potential difference induced by sodium sweat content, this analysis demonstrates the feasibility of a piezoelectric sweat sensor for continuous noninvasive sodium monitoring. Since piezoelectric materials have never been used evaluated for this field of biomedical sensing, suitable materials were never established. This examination effectively compared two distinct materials, PZT and PVDF. Through an acceleration analysis, it is clear that despite the flexibility and biocompatibility of PVDF, it was unable to differentiate between frequencies induced by external excitations and applied voltage [34]. Therefore, PZT has the greatest potential for PZT future sweat sensing applications. Despite drawbacks of PZT, such as natural brittleness, the low potential differences applied by sweat do not cause significant stress on the material, rendering it feasible [35], [36]. By addressing limitations of wearable sensor technologies, this experiment determined the effects of skin temperature on sensor output. Additionally, the necessity of pilocarpine has been alleviated since this sensor operates effectively under low volumes and sweat rates [37], [38]. This allows for continuous sodium monitoring without potential skin burns, hence providing athletes with a viable method for early diagnosis of potentially lethal disorders.

**Future Investigations**

While this experiment evaluated sensor accuracy as volume increases, a single sodium concentration was used. When varying sweat concentrations are evaluated, there may be inaccuracies in acceleration due to leftover salt deposits following evaporation of sweat. A polyvinyl chloride (PVC) sodium selective membrane can be integrated into the sweat pad [39]. This can result in sodium transport towards the skin, allowing for natural reabsorption. Further, the sensor must also be optimized to sense other analytes present in perspiration, such as potassium, glucose, and magnesium. Current methods utilize multiple ion specific electrodes in order to differentiate between each electrolyte [40]. However, this would force the use of multiple beams, which is not cost effective. Therefore, future investigations
should be directed toward finding differences in the actuation of PZT for sodium versus other electrolytes, which may be influenced by their specific resistivity [41].

Miniaturization of piezoelectric sensors is beneficial since they become portable and lightweight [42]. This can be accomplished by replacing the scanning vibrometer with a microelectromechanical system (MEMS) accelerometer [43]. While this can lead to significant reduction in sensor size and sensor cost, the mass of the accelerometer can provide another source of acceleration on the PZT beam [44]. Furthermore, the orientation of the accelerometer with regard to physical human movement is unpredictable, which may prevent accurate estimation of sodium concentrations [45]. Resolving this limitation will enable sophisticated electronic functionalities to be integrated, such as a transmitter, so that information regarding health status can be delivered directly to the user’s cellular device or computer.

Applications

Addressing these specific aspects is a fundamental step in preparing a product for widespread consumer usage. The PZT sweat sensor can then be employed to continuously monitor sodium levels in athletes so that the general population can realize when they are at risk of hyponatremia prior to the onset of potentially lethal afflictions. The key disorders that can be prevented are arrhythmias and TBI [46]. Furthermore, TBI has been associated with a wealth of neurological complications, such as syndrome of inappropriate antidiuretic hormone (SIADH), cerebral salt wasting syndrome (CSWS), and central diabetes insipidus (CDI), all of which can further induce hyponatremia and irreversible damage to an athlete [47]–[50]. The ability for piezoelectric materials to deform due to electrolytic conductivity is a breakthrough in the field of piezoelectric sensing. This innovative concept should not be limited solely to athlete sweat monitors. The PZT sweat sensor can be cross applied to monitor sodium deficiencies in over eight million anorexia and bulimia nervosa patients, since eating are correlated with sodium imbalances [51], [52]. Such applications could not have been realized by past sensors due to their inherent limitations. Therefore, taking advantage of this platform is imperative to facilitate real time physiological evaluations for understanding the health status of each individual in our society.
References


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