

Investigating the Use of Na₂SO₄ and MgSO₄ Electrolytic Solutions for Bulk Electrolysis Micro-Flowrate Pump

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ABSTRACT

A bulk energy-efficient electrolytic micro flowrate pump which generated the electrolytic gas to push syringe to inject the medicine are investigated in this paper. Two electrolytic solutions: Na₂SO₄ and MgSO₄, are electrolyzed to use as the power resource for pump. The results indicated that the preparation time of syringe injection is shorter with MgSO₄ electrolytic solution because of its lower impedance, as well as shortened entire operation time for the pump. However, the results also showed that it would be more difficult to control the process of electrolysis with MgSO₄ electrolytic solution because it easily yield white MgO precipitants in electrolysis, which caused unstable syringe injection flow rate, and decreased operating efficiency of pump. On the other hand, when using Na₂SO₄ electrolytic solution with higher impedance, it will cause longer preparation region and entire operation time for the pump. Due to there's no precipitants yielded from the process of electrolysis, thus its syringe injection flow rate would be stable and controllable, as well as higher operating efficiency. In addition, experimental result showed that a higher electrolytic current would result a higher syringe injection flowrate, and increase the operating efficiency for electrolytic pump.

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INTRODUCTION

Application of micro flow pump to medicine injection which could maintain the optimal medicine concentration at an appropriate time to obtain the best result of treatment and reduce the side effects of traditional oral intake and injection medicines has gradually improved in recent years. Micro flowrate pump can reduced the feelings of pain for patients, including cancers, chronic diseases, and cramps. In 1970s' the micro flow pump medicine injection has mainly used to control diabetes patients' blood glucose, and often applied to the animal studies and clinical researches. Micro flowrate pump can be divided into two major types: mechanical and non-mechanical. The mechanical-driven type adopts external actuated device to make liquid inside pump to be transported toward designated direction; and non-mechanical type is driven by electric current, electromagnetic, phase change or electrochemistry principles to achieve a lower flow rate.

Therefore, micro flowrate pumps can be applied to many disciplines, such as chemical analysis of medicine and industrial-process control (Zengerle et al.,1992), potential of biocompatibility (Neagu et al., 2000; Wang et al., 2005; Lindholm et al., 2004; Takami et al., 1997) applications of chemistry, medicine and biomedicine (Olsson, et al.,1997; Nisar, et al., 2008; Amirouche, et al., 2009). The micro flowrate pump systems include two major parts: actuating chamber and valve. Actuating chamber contains a chamber with vibrating thin-film in reciprocating cycle to drive the liquid operation. In addition, according to its different actuating methods, it can be divided into the pizeoelectric, electrostatic, thermo, electromagnetic and shape memory alloy methods. Besides, the valve part can drive the liquid to flow toward single direction. Zengerle and Ulrich (1995) developed the electrostatic micro flow pump, its features are using the electrostatic force to make thin-film vibration motion to drive the fluid. The dimension in this experiment is 7mm×7mm×2mm, within the drive voltage 200 V, the maximum flow rate is 150μl

min. Kim et al. (2004) and the research team initiated the piezoelectric micro pump, the structure of pump elements is mainly made of dimethylsiloxane silicone, by using SU-8 as the master model to make duplication. In addition, the valve structure is a dual nozzle-diffuser, thus it won't have any damage and fatigue problems. Such pump can react quickly and has a larger flow rate for liquid. In their research, the maximum back pressure will be 175kPa when its voltage is 150V, and its flow rate can reach 32.9 $\mu\text{l}/\text{min}$ when its driving frequency is 300 Hz. Jeong and Yang (2000) proposed the thermos-micro flow pump. One of its characteristics is used the ripple shape P+ silicone thin-film, since the deformation quantity is comparatively bigger than thin-film and the pump showed better performance. Dimension of thermo micro flow pump's ripple P+ silicone thin-film is 4mm \times 4mm \times 2 μm , and the maximum deformation quantity at the center is 100 μm . The maximum flow rate is 14 $\mu\text{l}/\text{min}$ when the input voltage is 8V and frequency is 4 Hz. Berg et al. (2003) used PDMS to design a discrete, two-stage peristaltic micropump and brought out the gas-driven micro flow pump, which is mainly composed of gas chamber, conduit and flow channel. Its driving part is mainly based on thin-film. The active valve is used as the main element to control the fluid direction. In addition, its gas-driven actuation is used nitrogen as the pneumatic supply for pump pressure. Its main structure is adopted dimethylsiloxane silicone as the main of elements, which can achieve the maximum flow rate of 0.098~0.117 $\mu\text{l}/\text{sec}$. Yamahata et al.(2005) developed the electromagnetic micro flow pump. This pump is mainly composed of 4 acrylic sheets, and used the repetitive motion of Neodymium (NdFeB) magnet as the power supply for pump. Besides, mixed magnetic powder with PDMS to make the permeable thin-film, in order to enhance the magnetic force and pump driving performance. When the driving frequency is under 12 Hz, micro flow pump can reach the maximum flow rate 0.4 ml/min, and driving force can reach 120 Pa.

In recent years, in order to increase the driving force and lower the energy consumption, electrolysis pump was developed for many applications. Xie et al. (2004) presented that they used the electrospray ionization ion trap mass spectrometry to integrate electrolysis-based micro flow pumps to make a single-chip system on the micro-hybrid electrospray ionization nozzle. Cr-Au electrode is used for the pump, and the electrolytic pump design is adopted 2 pump chambers and a hybrid chamber, by means of electrolysis to push the liquid to the hybrid chamber, and thus its pumping flow rate is around 2.3ml/min. Chan et al. (2010) also developed an electrolysis micro pump, including a microfluidic chamber structure and micro-electrode. It has assembled on glass substrate by using PDMS, and the electrode was deposited in closed microfluidic channel. The main purpose of this

pump is to apply to the heat dissipation for electronic products. In their experiment, AC/DC current were applied to the gap between electrodes to conduct the testing. When applying AC current to the electrode, bubbles generated from electrolysis reaction, and they will generate hydrogen and oxygen interactively on a same electrode. In addition, there're 3 testing sizes: 20 μm , 40 μm and 60 μm , for the gap between 2 electrodes. The result showed that when the distance between 2 electrodes is 20 μm , its flow rate will be higher than 40 μm and 60 μm , which proved that the in-between of these electrodes will affect on the electrolysis rate, thus for the electrolysis driven micro pump, its maximum flow rate is 37.8 $\mu\text{l}/\text{min}$. Li et al. (2010) studied a electrolysis pump with electrochemistry driving with membrane vibration. This electrochemistry pump include: PDMS structure with high flexibility can separate electrochemistry reaction from solution; improved design of electrolysis electrode to speed up the bubbles generation; long-term wireless control can reduce power consumption; and use biocompatible materials for bio-applications. Therefore, it has developed the implantable medicines transmission micro pump, within 1mA current, its flow rate can reach 6.5 $\mu\text{l}/\text{min}$. Gensler et al. (2010) also developed an implantable medicine transmission device. It contained an electrochemistry tank, refillable medicine storage tank, and double regulating valve. This research team made two types of pump, one is an active pump, and another is a passive pump. Active pump is used external force to push the liquid inside the pump outward, and passive pump is used diffusion principle to make the liquid inside the pump to flow from high concentration to low concentration. It can conduct the direct medicine intake from the source of organism's diseases in order to achieve the cure function. Lee et al. (2010) designed the driving mechanism of electrochemistry actuator based on the reversible electrolysis process of water. Compare results of the measurement and simulation for the electrochemistry actuator, electrodes were used by Silver Chloride (AgCl) and Platinum (Pt).

It was found that electrolysis-yielded bubbles attached to the electrode would reduce effective flow of electrons between electrodes, as well as decrease the efficiency for electrochemistry actuator. Besides, the injection flow rate is dependent on the gas generation rate and the flow resistance output into channels. Experimental result proved that electrode coated with Pt can be conductive to improve the reversible reaction of electrolysis, and electrolysis chambers' Pt electrode can become the catalyst for hydrogen's reversible reaction. Sheybani et al. (2011) announced that they used electrodes coated with Nafion to make the electrochemistry actuator, and adopted the wireless driving method to produce a high-efficient electrochemistry pump. They conducted electrolysis to the coated Nafion and coated Pt-Ti

electrodes, and compared their flow rate and pump efficiency. Then the experimental results showed that Pt-Ti electrode with Nafion coating can rapidly diffuse bubbles far away electrode's surface, which caused electrolysis to increase efficiency, and its flow rate can reach 141 μ l/min within current 13mA. Moreover, Pagonis et al. (2012) developed a new actuator which is based on the use of electrolytic water in microfluidic system pump. They used printed circuit board (PCB) to make electrolytic electrode and micro channel, and fabricated pump chamber on PCB directly. Then, gas produced from the closed structure during the process of electrolysis which resulted in a pressure increase to make liquid to flow into micro channels. Its flow rate range is between 20 μ l/min to 135 μ l/min.

In previous researches, electrolysis driving method had been proved to be a successful technique for microfluid application et al. (1988). Electrolysis is a process of using electrolytic solution to generate oxygen and hydrogen that can result in a huge volume expansion to push the fluid flow. The advantages of electrolysis pump are lower power consumption, more precise flowrate control and larger driving force (Li et al., 2010). Besides, electrolytic pump also has an advantage of simple mechanical design, it only needs one chamber and 2 electrodes, which can be easily passed the design flexibility to realize the design of such driver in micro flowrate control. However, there are very few studies in portable bulk electrolytic pump for micro flowrate medical injection applications. Therefore, in order to understand the flow phenomena and possibility design of bulk electrolytic pump for medicine injection applications, we realize the portable bulk energy-efficient pump that is adopted electrolysis driving method and we use biocompatible liquids (Na₂SO₄ and MgSO₄) to be the electrolytic liquids for our bulk electrolytic micro-flowrate pump.

ELECTROLYSIS MODEL

This study is mainly used the electrochemistry reaction to make micro flow pump, and utilized the electrolysis to generate hydrogen and oxygen. The resulting gas volume (hydrogen and oxygen) would be enlarger than water volume for several times. This larger gas volume can be calculated using the total reaction of electrolysis, and its equation is as follows:



From aforesaid chemical equation, it showed that with 1 mole of water electrolysis, it will generate the 2 mole of hydrogen gas and 1 mole of oxygen gas. Besides, the pressure and flowrate of gas generated from electrolysis that can be hypothesized according to Faraday's Law and Ideal Gas Law are as following (Chan, et al., 2010):

$$Q = \frac{dV}{dt} = \frac{IRT}{FPz} \quad (2)$$

$$P = \frac{nRT}{V} = \frac{ItRT}{zFV} \quad (3)$$

where, Q is flow rate, P is pressure, n is the moles of gas, R is the constant number of ideal gas, T is temperature, V is volume, I is electrolytic current, t is the powering time, z is the number of electrons, F is Faraday constant (9.649×10^4 C/mol). In the hypothesis of constant temperature and atmospheric pressure, the volume of generated gas is proportional to the applied current.

This electrolysis pump can generate gas (2 moles of hydrogen gas and 1 mole of oxygen gas from 1 mole of water electrolysis) to push the syringe and then the liquid or drugs can be delivered from the syringe; therefore, the volume of gas generated and its corresponding flow rate of liquid would depend on applied electrolytic current directly, thus the constant current control would be the priority of our control methods. Within the gas pressure controlled by applied electrolytic current, the theoretical pump flow rate ($q_{\text{theoretical}}$ m³/s) can be showed as the following Equation:

$$q_{\text{theoretical}} = \frac{3I}{4F} V_m \quad (4)$$

where I is electrolytic current (A); F is Faraday constant; V_m is the mole gas volume at NTP conditions (25 $^{\circ}$ C and 1 atmospheric pressure), and its volume size is 24.5×10^{-3} m³/mol. Its theoretical gas volume ($V_{\text{theoretical}}$, m³) equation is as following:

$$V_{\text{theoretical}} = q_{\text{theoretical}} t = \frac{3I}{4F} V_m t \quad (5)$$

where t is the total operation time, and its unit is second (s). Therefore, electrolysis's driving pump efficiency (η) can be defined as follows:

$$\eta = \frac{V_{\text{experimental}}}{V_{\text{theoretical}}} \quad (6)$$

where $V_{\text{experimental}}$ is obtained from experimental gas volume.

EXPERIMENT

Electrolytic micro flow pump is composed of three major parts: 1. Constant current control circuit; 2. Power transmission mechanism; and 3. Syringe and the design mechanism and circuit are shown in Fig. 1. Since the current control is the key factor of electrolytic micro flowrate pump to control the generation rate of electrolytic gas. If stable current can be provided and then it can maintain pump's stable pushing force. Therefore, constant current circuit is adopted in this experiment. Circuits contained capacitance, transistor LM317, variable resistance and digital display for current and voltage. Related circuit diagram is as shown in Figure 1(a). Besides, in our experiment, the variable electrical current was applied to control the gas's generation rate which can increase gas-generated pressure to push the double acting

pneumatic cylinder. This pneumatic cylinder moving can push a small type syringe to make the injection process. Besides, in order to avoid the corrosion of electrode pads during electrolysis process, the Pt was used to be the electrical pads. Because pump is driven by gas, thus the airtightness is pretty important factor of pump chambers. Therefore, in our design, a rubber O-ring is placed on the center at the connection between pump's main body and upper cap, and applied airtight connector in the gas transmission process.

This electrolytic micro flow pump is utilized the pneumatic cylinder to push small type syringe to make injection action, and then used syringe to push or squeeze liquid with micro flowrate transmission into human body for long term injection. In this experiment, Luer Lock syringe, with 3ml of volume, is used as the media device of transmitting simulation medicines. Besides, in order to avoid unstable injection flow rate caused by shaking and deviation of pneumatic cylinder and syringe as conduct injection, as a result, we additionally set a secure base to fix syringe and pneumatic cylinder, which would provide support and fixation when squeezing syringe. Furthermore, for conveniently observing the operation situation for small type syringe; therefore, the fix bases were made by the transparent acrylic.

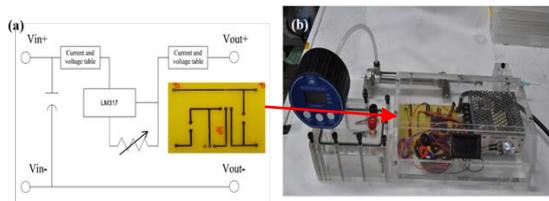


Figure 1 Constant-current control circuit design and pump assembly (a) Circuit design diagram (b) Actual pump assembly.

RESULTS AND DISCUSSIONS

After electrolytic micro flowrate pump assembled, pneumatic cylinder is the major actuating element which connected with the main body of this electrolytic micro flow pump. Its purpose is to convert the gas pressure into pushing force, and then it can squeeze out the liquid from a syringe. In our experiment, biocompatible $MgSO_4$ and Na_2SO_4 are selected as electrolytic solutions and its electrolytic gas provide the driving force for syringe injection. Besides, in this paper, 10wt% physiological saline is adopted as syringe injection liquid and its coefficient of viscosity is 0.9cP in room temperature $25^\circ C$.

Figure 2 presented the experimental operation diagram for pressure and time. It showed that pressure will be increased with time increased, and pressure approached to a constant value when reach a certain time. Because this study adopted electrochemistry as the driving method, after applying the electrolytic

current at the initial stage, electrolysis tank started to generate gas. The pressure generated by these gases but it couldn't provide enough pushing force to push syringe at preparation region (the piston of pneumatic cylinder doesn't move); that is, the driving force from the gas generation still behind the static friction of syringe piston, syringe is not yet activated. As a result, gas generation in this region will cause pressure to show a linear increase trend. In addition, according to the electrochemistry theory, as shown in Equation (3), increased electrolytic current will enhance its electrolytic reaction rate, and directly caused more electrolysis gases, which increase the pressure rapidly. Thus, when gas has increased to a certain level, as well as its gas force became larger than the static friction force of syringe, and then syringe's internal piston can be pushed and started the operation of medicines injection. As a result, we can divide the delivery process into two regions: preparation region and injection region, where the preparation region is defined as the period that the force of pushing piston has not yet achieved the maximum static friction force, thus the piston has not yet activated, and its flowrate is 0. On the contrary, as shown in Fig. 2 & 3, the injection region is defined as the period that the force of pushing piston has exceeded the maximum static friction; therefore, the piston has started actuating and making medicines injection. Fig. 2(b) showed the correlation diagram of pressure vs. time when the electrolytic solution is Na_2SO_4 and electrolytic current controlled between 50-100mA. This figure also displayed that when electrolytic current has increased, its gas generation rate and pressure increasing rate will be increased as well. As using Na_2SO_4 as the electrolytic solution, electrolytic current control has increased from 50mA to 100mA, and its pressure increasing rate will be increased from 1.15kPa/min to 2.25kPa/min in the preparation region, respectively. In addition, when electrolytic current is 50mA, and the time is more than 31.6 minutes, then the internal gas pressure could achieve to actuate the pneumatic cylinder to push the syringe. There exists a volume gap due to pistons inside acting pneumatic cylinder and syringe actuating forward, thus such volume gap needs to be filled up by electrolysis gas, and then syringe can start to continuously actuating in injection region. However, at the beginning of injection region, when its piston just conquered static friction force of syringe wall, thus the force generated of electrolytic gas would be greater than dynamical friction because static friction is greater than dynamical friction; therefore, the gas generation speed of electrolysis at initial of injection region would be faster than the moving speed of pneumatic cylinder, which could make slight increase of pressure after pushed it forward, and then it will reach a stable state in a short time. This could make internal and external pressures of electrolysis pump to be constant, and its slope would tend to approach horizontal. In addition, when electrolytic current is

greater than 90mA, due to electrolytic current is larger and generation rate of electrolytic gas is too fast, which could rapidly squeeze liquid out of syringe and its injection region time is comparatively shorter. This result also showed that if adopting electrolytic pump injection, its electrolytic current needs to be controlled within suitable operating range in order to precisely control the flow rate of medicine injection.

Fig.2 (b) showed the electrolytic solution is MgSO₄ and the diagram of pressure and time when controlling electrolytic current between 50-100mA. Due to electrolytic pump needs to generate a certain pushing force to push syringe to make the injection, thus the corresponding process also should be divided into preparation region and injection region. In preparation region, as the result of Na₂SO₄ electrolytic solution, the increasing rate of its pressure will be increased along with the increase of electrolytic current. However, it is worth to mention, when using MgSO₄ electrolytic solution, the pressure's increase is not stable. The reason is that white MgO will be produced on electrode when MgSO₄ is electrolyzed. It will cause unstable generation rate of electrolytic gas. Moreover, when electrolytic current is between 80mA and 100mA, the reason of rapid decrease for the pressure at the end of experiment is that the electrolytic gas pressure inside pneumatic cylinder is far greater than required pushing pressure, and resulted in exceeded pushing force to rapidly squeeze out the liquid from syringe, and then the pressure decreases rapidly.

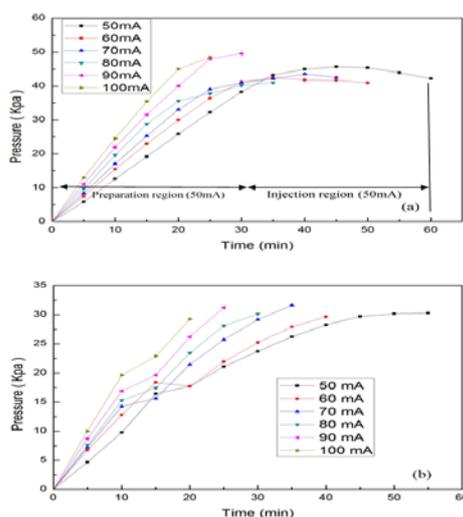


Figure 2 Correlation diagram of pressure and time with (a) Na₂SO₄ (b) MgSO₄ electrolytic solution and (a) showed the definition of preparation region and injection region at applied electrical current at 50mA.

Fig. 3 showed the relationship between volume and time for the fluid that pushed the syringe by the electrolysis gas from different electrolytic solutions: Na₂SO₄ and MgSO₄. This figure also displayed that the electrolytic pump's delivery process also divided into preparation region and injection region. As

showed in Fig. 3(a), when electrolytic solution is Na₂SO₄ and its electrolytic current is 50mA, due to the pressure generated from electrolytic gas is still not enough to push the piston inside syringe, thus when time is less than 31.6 minutes, the pumped volume is 0.

This is the preparation region abovementioned, the pressure generated by its electrolytic gas that is mainly to conquer the static friction between acting pneumatic cylinder and syringe; therefore, there still has no liquid that ejected from syringe. However, when the force generated by electrolytic gas is greater than static friction force, the liquid will be squeezed out gradually, and the ejected liquid volume showed a nonlinear change at the initial pushing stage. At the time more than 40 minutes, force generated by electrolytic gas and piston's resistance will achieve balance, and then ejected liquid volume will show a linear increase with time. The Figure 3(a) also showed that when electrolytic current increasing, the curve slopes in injection region is increasing; that indicated when electrolytic current is increased, the flow rate of ejected liquid from syringe is increased. When electrolytic current is increased from 50mA to 100mA, the flow rate of ejected liquid from the syringe will be increased from 1.85 μ l/sec to 7.32 μ l/sec. Fig. 3(b) showed the correlation diagram of pumped volume and time when using MgSO₄ as the electrolytic pump for electrolytic solution. Different from using Na₂SO₄ electrolytic solution, the use of MgSO₄ electrolytic solution will shorten the time of preparation region, and the ejected liquid volume will display a nonlinear change easily. Fig. 4 displayed the impedance diagram of Na₂SO₄ and MgSO₄ electrolytic solution, in such figure it obviously discovered that only 3.22V voltage required when using MgSO₄ electrolytic solution can achieve electrolytic current 50mA required in this experiment, but it needs 3.41V when using Na₂SO₄ electrolytic solution. It found that the slope of V-I diagram when using MgSO₄ electrolytic solution is higher than the one with using Na₂SO₄ electrolytic solution. Based on Ohm's Law ($R=V/I$), the resistance value of MgSO₄ electrolytic solution is 64.4 Ω , and Na₂SO₄ electrolytic solution is 68.2 Ω . As a result, it showed that the electrical conductivity of electrolytic solution prepared by using MgSO₄ is higher than the one prepared by using Na₂SO₄. In another word, as showed in Fig. 5, the preparing time of syringe piston for MgSO₄ electrolytic solution is faster than using Na₂SO₄ electrolytic solution. Therefore, with the same electrolytic current, the impedance of MgSO₄ electrolytic solution is lower than Na₂SO₄ electrolytic solution comparatively, which made the faster electrochemistry reaction with MgSO₄ electrolytic solution in pump chamber. In addition, it can produce more hydrogen and oxygen. Within the same time, the time required by preparing MgSO₄ electrolytic solution will be comparatively shorter than Na₂SO₄ electrolytic solution. Therefore, the use of MgSO₄

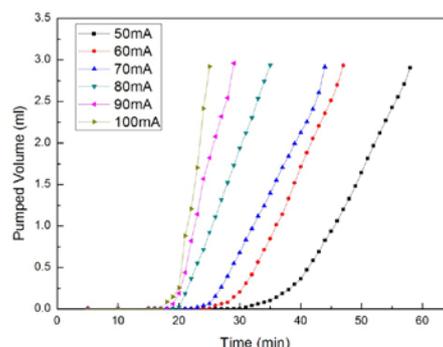
electrolytic solution can greatly shorten the time of preparation region.

Nevertheless, MgSO₄ electrolytic solution can shorten the time of preparation region, but due to MgSO₄ electrolytic solution would be easily produced white precipitate, such as MgO, when it is electrolyzed, thus it could cause unstable generation rate of electrolytic gas, as well as unstable gas pressure. As a result, the liquid volume ejected from syringe didn't show a linear phenomenon as using MgSO₄ electrolytic solution, but showed an irregular shaking as shown in Fig. 3(b). Moreover, when electrolytic current is increased, due to MgSO₄ electrolytic solution has higher electrical conductivity, it can shorten the time of preparation region, but it also may cause unstable gas pressure, as well as made unstable ejected flow rate for syringe (as shown in Fig. 2 (b)); as a result, syringe's ejected liquid flow rate is not necessarily higher than using Na₂SO₄ electrolytic solution. Fig. 3(b) also showed that when using MgSO₄ electrolytic solution, electrolytic current will be increased from 50mA to 100mA, its ejected liquid flow rate of syringe will be also increased from 1.31 μ l/sec to 3.45 μ l/sec. Also, Fig. 2 (b) displayed that when using MgSO₄ electrolytic solution, the electrolytic current is greater than 80mA, syringe's liquid can be easily ejected. To summarize discussions in Fig. 2 and Fig. 3, for the flow rate stability of electrolytic micro flow pump designed in this study, the use of Na₂SO₄ electrolytic solution is better than using MgSO₄ electrolytic solution.

As shown in Figure 6, if considering total operation time (= preparation time + injection time) of this experiment, that is, it is the time of that the experiment started to electrolyze in pneumatic cylinder to react with syringe, until completely squeezed the liquid out of syringe. We can discover that the increase of electrolytic current could shorten total operation time for the pump. In Na₂SO₄ electrolytic solution, the experimental time of 58 minutes for current 50mA has shortened to the experimental time of 28 minutes for current 100mA. However, in MgSO₄ electrolytic solution, the total operation time has shortened from 55 minutes for current 50mA to 26 minutes for current 100mA. Such result showed that it has a lower impedance when using MgSO₄ electrolytic solution, the electrolytic speed of MgSO₄ electrolytic solution will be faster than using Na₂SO₄ electrolytic solution. Therefore, the use of MgSO₄ electrolytic solution with higher electrolysis efficiency will shorten its total operation time and make the difference between experimental time and theoretical time smaller.

Figure 7 showed the comparison diagram of ejected flow rate and theoretical flow rate by two different electrolytic solutions Na₂SO₄ and MgSO₄ in electrolytic pump syringe. The syringe flow rate that is calculated the ratio of syringe volume to actuating time. In addition, the theoretical flow rate is calculated

from above mentioned Equation (4). Results showed that the operating current is between 50mA to 100mA when using Na₂SO₄ electrolytic solution, thus its flow rate is between 1.85 μ l/sec to 7.32 μ l/sec. μ l/s; when using MgSO₄ electrolytic solution the flow rate is between 1.31~3.45 μ l/s when current is between 50mA to 100mA. Even in Fig. 4, it showed that the impedance when using MgSO₄ electrolytic solution is lower than using Na₂SO₄ electrolytic solution, and the starting time of syringe with using MgSO₄ electrolytic solution is faster (preparation region is shorter). However, since white precipitate, such as MgO, will be easily produced in the electrolysis of MgSO₄ electrolytic solution; therefore, the generation rate of electrolytic gas is unstable and cause unstable flow rate when syringe ejected liquid in injection region; as a result, it resulted in an increase of injection region time. Lower syringe ejected flow rate is caused the syringe injection flow rate when using MgSO₄ electrolytic solution than using Na₂SO₄ electrolytic solution. Such result also indicated in Fig. 3, the correlation diagram of injection volume and time. Besides, Fig. 7 also showed that there's a certain difference between actual flow rate and theoretical flow rate. The reason why occurring this difference is that the power mechanism design of electrolytic micro flow pump used pneumatic cylinder to push syringe, there're pistons inside these two parts, electrolytic gas generated pressure needs to conquer the friction between piston and syringe wall to push syringe, and then resulted in a certain difference between actual flow rate and theoretical flow rate. Experimental result also discovered that when using Na₂SO₄ or MgSO₄ electrolytic solution, the electrolytic current increased with a rapid electrolytic reaction that will make actual flow rate to be more closed to theoretical flow rate. In addition, results in Fig. 7 also showed that white precipitate would be generated when using MgSO₄ electrolytic solution which would cause unstable electrolysis, the increase value of actual flow rate followed the increase ratio of electrolytic current is far lower than using Na₂SO₄ electrolytic solution.



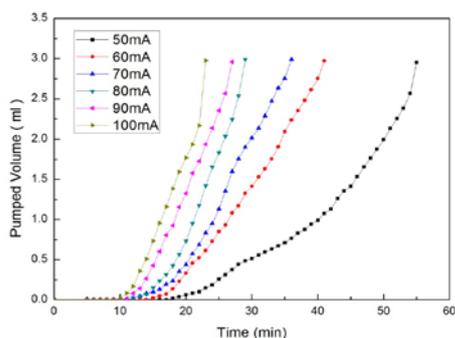


Figure 3 Correlation diagram of pumped volume and time with (a) Na₂SO₄ (b) MgSO₄ electrolytic solution and (a) showed the preparation region and injection region at applied electrical current at 50mA.

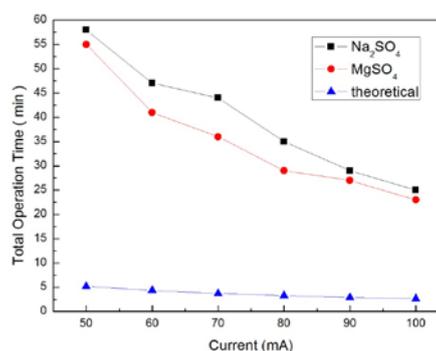


Figure 6 Correlation diagram of total operation times and electrolytic currents for Na₂SO₄ and MgSO₄ electrolytic solution.

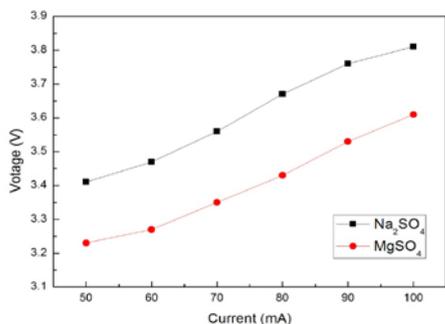


Figure 4 Correlation diagram of applied electrolytic voltage and electrolytic current with Na₂SO₄ and MgSO₄ electrolytic solution.

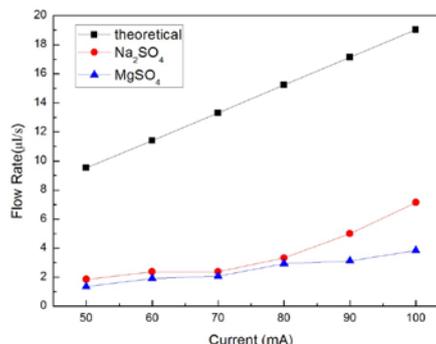


Figure 7 Correlation diagram of experimental and theoretical flow rates with different electrolytic currents for Na₂SO₄ and MgSO₄ electrolytic solution.

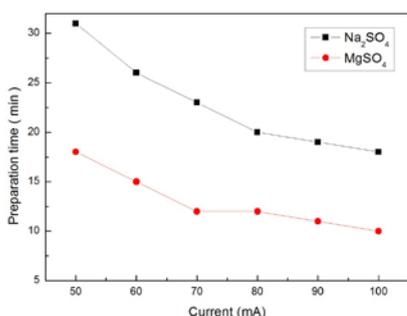


Figure 5 Correlation diagram of preparation times and electrolytic currents for Na₂SO₄ and MgSO₄ electrolytic solution.

In Figure 8, it showed the comparison diagram of driving pump efficiency for these two electrolytic solutions. Results displayed that the pump efficiency is lower when the current value is between 50mA to 80mA with using Na₂SO₄ electrolytic solution. It has the most stable flow rate at 3.5μl/s; in addition, when current reached 90mA and 100mA, its driving pump efficiency will be greatly increased, the reason is due to it has a higher electrolysis efficiency. However, as shown in Fig. 3, syringe's ejected flow rate would show a rapid increase, and the reason is that when piston starts actuating, dynamic friction between piston and syringe wall will be lower than static friction, in the meantime, the pushing force from the internal pressure is greatly larger than piston's dynamic friction force, which accelerate the piston moving by electrolytic gas pressure. With using MgSO₄ electrolytic solution, because it is affected by MgO precipitate, its syringe ejected flow rate would then be displayed an unstable state, and also greatly decreased its electrolysis efficiency. Therefore, the use of Na₂SO₄ electrolytic solution would increase its preparation time of syringe injection. However, with considering stability and driving pump efficiency, Na₂SO₄ electrolytic solution is the optimal selection

when electrolytic current is between 50mA to 80mA and it will have a stable and more easily controlled ejected flow rate for syringe.

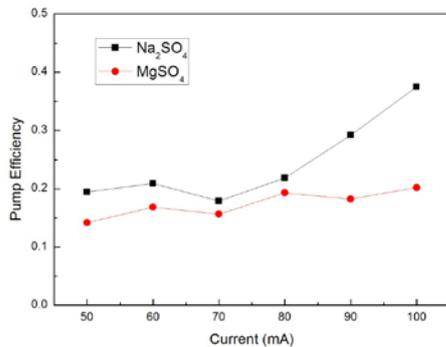


Figure 8 Correlation diagram of pump efficiency and electrolytic current in Na₂SO₄ and MgSO₄ electrolytic solution experiments

CONCLUSIONS

This study designed an electrolytic micro flow pump with low power consumption. Two different biocompatible electrolytic solutions: Na₂SO₄ and MgSO₄ electrolytic solutions, were used to be the power resources for pump. Results showed that its impedance is lower with MgSO₄ electrolytic solution, thus it would have a shorter preparation time for syringe injection, as well as shortened the pump's overall operating time. If electrolytic current could be controlled between 50-100mA, the range of its syringe injection flow rate should be between 1.31~3.45 $\mu\text{l/s}$ with MgSO₄ electrolytic solution. When using Na₂SO₄ electrolytic solution, since it has higher electrolytic solution impedance, then the preparation time of syringe injection will be longer, as well as pump's total operating time. In addition, with same electrolytic current range (50-100mA), the syringe injection flow rate range is between 1.85~7.32 $\mu\text{l/s}$. However, the experimental result also showed that due to the white precipitate, such as MgO, will be produced when using MgSO₄ electrolytic solution. It will cause an unstable syringe injection flow rate, and decrease its syringe injection efficiency. In addition, excessive electrolytic current would be made syringe's injection flow rate too big, thus syringe's liquid would be ejected rapidly, which caused the syringe injection flow rate is difficult to control. Therefore, the optimal operating condition is using Na₂SO₄ as the electrolytic solution, and the most stable flow rate 3.5 $\mu\text{l/s}$ could be reached at current 80mA. It is important indicated that when using bulk electrolytic pump, its delivery process can be divided into preparation region and injection region. As a result, if the volume between electrolysis tank and pneumatic cylinder can be smaller, it can greatly reduce the time of preparation region. Therefore, if the micro-electromechanical process can be used to make micro-

scale electrolytic pump, it can greatly decrease the volume between electrolysis tank and compound pneumatic cylinder and then it can increase the pump efficiency and feasibility for micro flowrate medical injection applications.

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REFERENCES

- Amirouche, F., Zhou, Y., and Johnson, T., "Current micropump technologies and their biomedical applications", *Microsystem Technology*, Vol.15, pp.647-666 (2009).
- Berg J. M., Anderson R., Anaya M., Lahlouh B., Holtz M., and Dallas T., "A two stage discrete peristaltic micropump", *Sensor and Actuator*, Vol.104, pp.6-10 (2003).
- Chan, S.C., Chen, C.R., and Liu, C.H., "A bubble-activated micropump with high-frequency flow reversal." *Sensor and Actuators A*, Vol.163, pp.501-509 (2010).
- Gensler, H., Sheybani, R., Li, P.Y., Lo, R., Zhu, S., Yong, K.T., Roy, I., Prasad, P. N., Masood, R., Sinha, U. K., and Meng, E., "Implantable mems drug delivery device for cancer radiation reduction." *IEEE 23rd International Conference on Micro Electro Mechanical Systems (MEMS)*, pp.23-26 (2010).
- Jeong, O. C., and Yang, S. S., "Fabrication and test of a thermopneumatic micropump with a corrugated p+ diaphragm." *Sensors and Actuators A*, Vol.83, pp.249-255 (2000).
- Kim, J. H., Kang, C. J., and Kim, Y. S., "A disposable polydimethylsiloxane-based diffuser micropump actuated by piezoelectric-disc", *Microelectronic Engineering*, Vol. 7, pp.119-124 (2004).
- Lindholm, L., Westerberg, M., Bengtsson, A., Ekroth, R., Jensen, E., and Jeppsson, A., "A closed perfusion system with heparin coating and centrifugal pump improves cardiopulmonary bypass biocompatibility in elderly patients", *Ann. Thorac. Surg.*, Vol. 78, pp.2131-2138 (2004).
- Li, P. Y., Sheybani, R., Gutierrez, C. A., Kuo, J. T. W., and Meng, E., "A parylene bellows electrochemical actuator." *Journal of microelectromechanical systems*, Vol.19, pp.215-228 (2010).
- Lee, D.E., Soper, S. A., and Wang, W., "Fabrication and mathematical analysis of an electrochemical microactuator (ECM) using electrodes coated with platinum nanoparticles." *Microsyst Technol*, Vol. 16,

- pp.381–390 (2010).
- Meng, E., and Hoang, T., “MEMS-enabled implantable drug infusion pumps for laboratory animal research, preclinical, and clinical applications.” *Advanced Drug Delivery Reviews*, Vol. 64, pp.1628–1638 (2012).
- Neagu, C., Jansen, H., Gardeniers, H., and Elwenspoek M., “The electrolysis of water: An actuation principle for MEMS with a big opportunity.” *Mechatronics*, Vol. 10, pp. 571–581 (2000).
- Nisar, A., Afzulpurkar, N., Mahaisavariya, B., and Tuantranont A., “MEMS-based micropumps in drug delivery and biomedical applications”, *Sensors and Actuators B*, Vol.130, pp.917–942 (2008).
- Olsson, A., Enoksson, P., Stemme, G., and Stemme, E., “Micromachined flat-walled valveless diffuser pumps.” *Journal of Microelectromechanical Systems*, Vol. 6, pp. 161-166 (1997).
- Pagonis, D.N., Petropoulos, A., and Kaltsas, G., “A pumping actuator implemented on a PCB substrate by employing water electrolysis. *Microelectronic Engineering*, Vol. 95, pp.65–70 (2012).
- Sheybani, R. and Meng, E., “High efficiency wireless electrochemical actuators: Design, fabrication and characterization by electrochemical impedance spectroscopy.” *MEMS 2011, Cancun, MEXICO*, pp. 1233–1236 (2011).
- Takami Y., Nakazawa T., Makinouchi, K., Glueck, J., and Nose Y., “Biocompatibility of alumina ceramic and polyethylene as materials for pivot bearings of a centrifugal blood pump”, *Journal of Biomedical Materials Research*, Vol. 36 No.3, pp. 381-386 (1997).
- Wang C. H., and Lee G. B., “Automatic bio-sampling chips integrated with micro-pumps and micro-valves for disease detection”, *Biosensors and Bioelectronics*, Vol. 21, pp. 419–425 (2005).
- Xie, J., Miao, Y. N., Shih, J., He, Q., Liu, J., Tai, Y. C., and Lee, T. D., “An electrochemical pumping system for on-chip gradient generation” *Anal. Chem.*, Vol. 76, pp. 3756–3763 (2004).
- Yamahata, C., Lotto, C., Al-Assaf, E., and Gijs, A. M., “A PMMA valveless micropump using electromagnetic actuation.” *Microfluid Nanofluid*, Vol.1, pp. 197-207 (2005).
- Zengerle, R., Richter, A., and Sandmaier, H., “A micro membrane pump with electrostatic.” *Micro Electro Mechanical Systems* Vol. 92, pp. 4-7 (1992).
- Zengerle, R., and Ulrich, J., “A bi-directional silicon micropump.” *Sensors and Actuators A*. Vol. 50, pp.81-86 (1995).

NOMENCLATURE

F	Faraday constant (9.649×10 ⁴ C /mol)
I	electrolytic current
n	moles of gas
P	gas pressure
Q	flowrate
R	the constant number of ideal gas
T	temperature
t	total operation time
V	gas volume
V _m	the mole gas volume at NTP conditions (25°C and 1 atmospheric pressure)
V _{experimental}	experimental gas volume
z	the number of electrons
η	driving pump efficiency

電解式微流量幫浦 設計與開發

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摘要

本研究主要是設計出低耗能之電解式微流量幫浦，研究電解之方式，將產生的氣體推動複合式氣壓缸來推動針筒進行藥物注射之動作。研究中使用兩種不同電解液 Na_2SO_4 與 MgSO_4 電解做為幫浦之動力來源並討論其優劣性。結果顯示使用 MgSO_4 電解液其阻抗較低，因此具有較短的針筒注射準備時間，並縮短幫浦整體操作時間。但是 MgSO_4 電解液在電解時會產生白色生成物，較不容易去控制電解過程，造成針筒注射流量較不穩定，幫浦操作效率下降。然而若使用 Na_2SO_4 電解液時，雖然電解液阻抗較高，準備區與幫浦整體操作時間較長。但因為在電解過程沒有任何生成物產生，針筒注射流量穩定且容易控制，並有較高的操作效率。此外，實驗結果也顯示較大的通電電流將會產生較大的針筒注射流量，將會提高電解式幫浦的操作效率。