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Pressure-driven ballistic Kelvin's water dropper for energy harvesting

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Abstract

In this paper, we introduce a microfluidic based self-excited energy conversion system inspired by Kelvin's water dropper but driven by inertia instead of gravity. Two micro water jets are produced by forcing water through two micropores by overpressure. The jets break-up into microdroplets which are inductively charged by electrostatic gates. The droplets land on metal targets which they gradually charge up to high voltages. Targets and electrostatic gates are cross-connected in a way similar to Kelvin's water dropper. Application of pressure as driving force instead of gravity as in Kelvin's dropper allows for much higher energy densities. To prevent overcharging of the droplets by the inductive mechanism and consequent droplet loss by repulsion from the target as in Kelvin's water dropper, a voltage divider using inversely connected diodes was introduced in our system to control the charge induction providing self-limiting positive feedback by the diode characteristics. Maximal 18% energy conversion efficiency was obtained with the diode-gated system.

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Introduction

As one of the renewable energy conversion methods, microfluidic energy conversion is relatively less known to the public. Recently we demonstrated a new form of microfluidic energy conversion named as "ballistic", which is both an efficient and powerful method to generate electric power. We showed that with a single-jet system a maximal conversion efficiency of 48% could be obtained.[1] In this conversion method water is accelerated by pumping it through a micropore, where it forms a microjet breaking up into fast-moving (>10 m/s) charged droplets. Droplet kinetic energy is subsequently converted to electrical energy when the charged droplets decelerate in the electrical field that forms between membrane and a metal target. Like traditional electrostatic gate control in micro/nanofluidics[2, 3], in our original device we also employed an electrostatic gate to charge the droplets and obtained a stable and adjustable current generation that is independent of the membrane zeta potential. With a well-controlled applied voltage and proper design of the gate, theoretically the droplet charge density can be increased until the Rayleigh limit, so that the target potential required for high efficiency can be decreased to the order of daily-use range.[1] However, for an electrical energy conversion device an external voltage source is cumbersome to implement. Instead, a self-excited energy conversion system will be much more convenient for applications, as it is in many other self-excited energy devices that have been studied. [4-8] Here we report on such a self-excited device.

Lord Kelvin invented an electrostatic high voltage generator by water dripping in 1876, generally named the 'Kelvin water dropper'.[9] Two metal buckets with an opening in the bottom drip water. The falling water drops pass through hollow metal rings and are collected by two other metal buckets. The hollow metal rings through which the droplets pass are cross-connected with both bottom buckets (a setup as in figure 1b). As a result, any tiny charge on the water drops (e.g. a positive charge) that left the left-top bucket will charge the bottom left bucket and hence the right side ring. The metal ring on the right side will then attract more negative charges to the water drops on the right side, so that more net negative charges will be collected by the bottom right bucket and the negative voltage increases on the left ring. Hence, a positive feedback exists and the two downstream buckets will continue this charging process until voltage breakdown occurs between the two bottom buckets, or drops are repelled and fall outside of the buckets. The Kelvin water dropper is thus a self-excited positive feedback system that charges water droplets and delivers charged droplets at high electrical potential driven by gravity.[10] A first step towards a practical useful voltage generator based on Kelvin's water dropper was shown recently, when Marin et al. demonstrated a microfluidic version of Kelvin's water dropper using water in oil droplets.[11] Also a self-powered ionization device can be produced based on Kelvin's water dropper where ionization occurs at very low potentials by providing very low internal energy to the ions.[12] However, the Kelvin water dropper itself has not been further improved as energy harvesting device since the droplets are easily been overcharged inducing much deflection (loss) of droplets on energy collection. Besides, the generated voltage is far too high for practical applications, and the harvested currents are very small with regard to the amount of water consumed.

Previously we demonstrated the single microjet ballistic system discussed above. [1] In the present paper, we apply an electrostatic charge self-induction mechanism inspired by Kelvin's water dropper to this inertia-driven (ballistic) energy conversion system and show that the disadvantages of Kelvin's water dropper can be overcome. The droplet charge will be derived from the same inductive charging mechanism as in Kelvin's droplet generator, employing two separate systems and cross-connecting targets and induction rings. As already mentioned above, one of the disadvantages of Kelvin's water dropper is that it uses a positive feedback system that is inherently unstable since the droplet charge keeps increasing until the droplets are deflected from the target because the work (required for droplets landing) done by electrical force overcomes the gravitational force ($mgh < qU_{target}$), where m, g and h are droplet mass, the acceleration due to gravity and the bucket height, q the charge quantity in the droplets, and U_{target} the target voltage. In our ballistic system overcharging of droplets will also result in droplet repulsion from the target, when the droplet kinetic energy $\frac{1}{2} mv^2$ is smaller than the electrical energy qU_{target} which the droplets would deliver when they reach the target ($\frac{1}{2} mv^2 < qU_{target}$). Here m, v, q are mass, velocity and charge quantity in droplets, and U_{target} is target voltage. The deflection of these overcharged droplets causes a lower efficiency.[1] To overcome the disadvantages of Kelvin's positive feedback mechanism, we therefore introduce voltage dividers (including an Ohmic and a non-Ohmic divider by reversely connecting diodes) in the system to properly control the induction voltages thus enabling a stable current generation and energy conversion as a self-gated system.

Ballistic Kelvin's water dropper – principle and setup

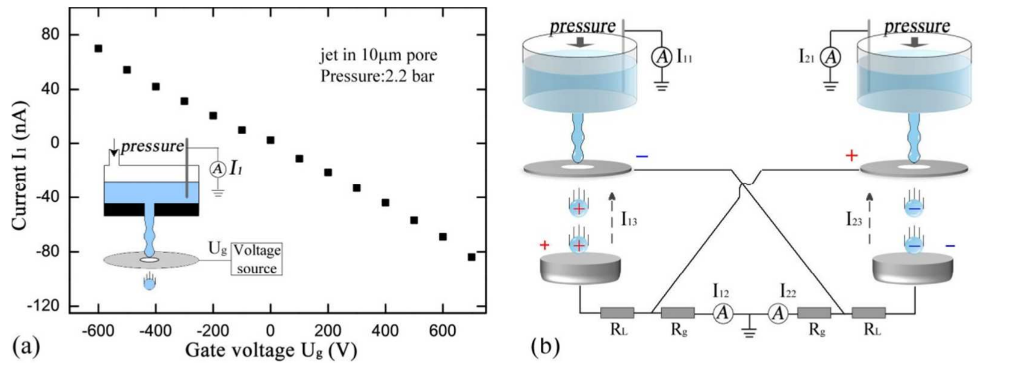

Figure 1. *a. The principle of controlling the upstream current by gate induction. The polarity of current I_1 can be controlled by applying a proper gate voltage. Solution: 0.1M KCl, pore diameter: 10 μm, applied pressure: 2.2 bar. b. Our self-excited ballistic energy system driven by pressure. Resistors form a voltage divider to separate the gate voltage from the target voltage.*

As we described previously, the energy conversion in a ballistic system can be influenced by using an induction ring to control the charge density of the droplets and hence the current carried by the droplets. A negative applied voltage on the gate (induction) ring thereby induces a positive current I_1 in the upstream reservoir. To investigate the possibility of inducing both a positive and negative current by gate induction, as needed for self-excited operation, we performed a preliminary experiment (setup shown in the inset figure 1a). As shown, the current can be tuned from positive (75nA) to negative (-83nA) by adjusting the applied gate voltage from -600V to 650V with a power source (Keithley 2410). The electrostatic induction of charges in the liquid jet is a capacitive process, and the resulting charge quantity in the droplets can be calculated as $q=C_{ind}*U_{target}$, where q and C_{ind} are charge quantities in the droplets and the capacitance of the induction gate, which is a function of the induction ring dimension and its distance from the jet.[13] The induction charge (current) thus is expected to increase linearly with the applied voltage as indeed is observed in figure 1a. Hence, we can conclude that adjusting the voltage on the gate ring enables not only quantities but also polarities of the droplet charge density. This provides a possibility to use the design of the Kelvin water dropper to build a self-excited ballistic energy conversion device.

Figure 1b shows a schematic picture of our ballistic Kelvin's water dropper energy conversion device. Two silicon nitride membranes with a single cylindrical pore with a diameter of 30 μm were mounted in pressurized PMMA reservoirs with rubber O-rings. The reservoirs contained degassed ultrapure 10m KCl solution. A layer of 150nm thickness Pt was sputtered at the back side of the chip for electrical connection with instruments via four metal pins. The mechanical input power can be calculated by measurement of the flow rate (Bronkhorst Cori-flow flow sensor) and pressure (Sensortechnics CTE 8016 GY7 pressure sensor). Two pieces of aluminum foil with a thickness of 0.2mm were attached under the chip holders to function as induction gate. Upstream currents are denoted as I_{11} and I_{21}, and the harvested currents as I_{12} and I_{22}. The currents, including the current generated by deflection of droplets from the target to the gate ring (I_{13} and I_{23}) can be measured by either a pico-ammeter (Keithley) or a multi-meter in voltage mode, showing the voltage difference over its integrated 10MOhm resistor induced by the current flow. Two stainless steel cups functioned as targets and were placed beneath the chip holders to collect charged droplets. The load resistance connected between these targets and ground was divided into two sections: resistors for electrical power generation and resistors for induction of the other jet, named R_L and R_g respectively. Resistors were immersed in an oil bath (Shell Diala S2-ZU-I) to prevent corona discharge and a layer of Delrin (Polyoxymethylene) was mounted at the edge of the cup for the same reason, at the location where the electrical field was expected to be the strongest.

Experimental Results

Kelvin's connection

In view of the successful generation of high voltage by Kelvin's water dropper, we first directly apply Kelvin's water dropper electrical connection in our ballistic energy conversion system (see figure 2a). For understanding of its functioning we can extract the electrical elements from the setup and make a simple circuit drawing as shown in figure 2b. Electrical currents I_{11} and I_{21} carried by the two jets were successfully generated with magnitudes up to 60 nA. The upstream currents from the two jets had opposite polarities, showing successful operation in current induction mode. However, almost no charged droplets were collected by the targets, as they were deflected to the gate as could be observed by the naked eye and was expressed in low values of the downstream collected currents I_{12} and I_{22}. These deflected droplets can be considered as a negative feedback to the gate voltage that keeps the droplet charge density at a proper level for stable operation. However, the deflection of droplets represents an energy loss source as it decreases the useful current through load resistance R_L. In addition, the deflected droplets contacting the gate electrodes decrease the current flow through R_g, thus doubling the electrical energy losses on R_g, since the total output energy can be calculated as $P_{out} = [(I_{12}+ I_{23})^2+(I_{22}+I_{13})^2]*R_L + (I_{12}^2+I_{22}^2)*R_g$. As expected, the weakness of this electrical setup is caused by the fact that the target is directly connected to the gate as in Kelvin's water dropper, and hence target voltage equals gate voltage. Gate voltages (and hence induced droplet charges) thereby become much higher than the ones needed for efficient operation. Thus, we introduced a voltage divider in the target circuit to independently control target voltage and gate voltage.

Resistor voltage divider connection

Two downstream circuits were investigated to perform self-induction. First, two 1TOhm resistors were series connected for power generation and gate induction (figure 2c). In figure 2d, we can see clearly that the upstream currents I_{11} and I_{21} increase rapidly after the upper electrodes were connected to the ammeter (the event is marked by a blue arrow). Deflected droplets accumulated at the bottom of the gate, dripping to the target, and causing the observed periodic transients in current level. Operation was still not optimal, since from figure 2e it can be seen that the downstream currents I_{12} and I_{22}, which are the currents through R_g, were still much smaller than the upstream currents, again due to partial deflection of the charged droplets.

As one of the most important performance characteristics of an energy conversion device, we calculate the efficiency which is defined as the ratio of electrical output power (P_{out}) to fluid mechanical input power (P_{in}), $eff = P_{out}/P_{in}$. We determine the electrical output power P_{out} by measuring the current (I_{i2}) flow through R_g and then deriving the current flowing through R_L by applying Kirchhoff's current law at the junction of R_g and R_L, stating that the current through R_L equals the sum of the current through the gate and R_g. The output power of the two jets thus calculated is 226µW. From measurements of the flow rate (13.1µL/s) and applied pressure (1.40bar), the efficiency of the resistive current divider system is calculated as 12.3%.

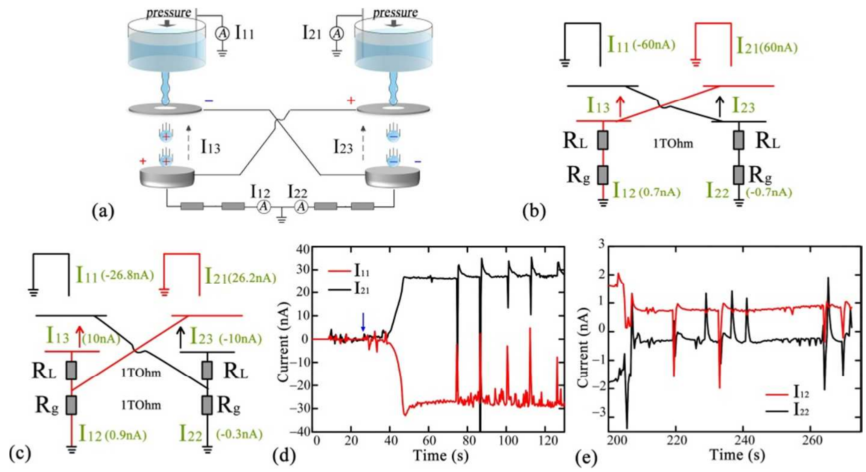

Figure 2. Measurements using the self-induction system. a. A schematical picture of the typical connection of Kelvin's water dropper. Induction rings are directly conncted to the bottom targets. b The connection scheme of Kelvin's water dropper. Most droplets are highly charged and are deflected to the gate ring. c. The connection scheme with voltage divider. d. Using the voltage divider, the upstream currents I_{11} and I_{21} increase as function of time. The blue arrow indicates when the induction gate was connected. d Using the voltage divider both downstream currents I_{12} and I_{22} are only a fraction of the upstream currents after some time of operation.

This efficiency is much lower than what we previously showed in a single jet with gate control via an external voltage source (48%)[3]. A possible explanation for this lower efficiency is that R_g is still too high for proper charge induction. As shown in Figure 1a, the induction charge increases linearly with gate voltage. For instance, assuming that no current loss occurs by deflected droplets, 10nA creates 10kV on the gate ring which is too high for current induction in our system according to our previous research (nearly -170V gate voltage was found to be optimal for R_L=1T Ohm). [1] Another reason for the low efficiency is that the highly charged droplets repel each other and form a cone when they move out of the gate, as was indeed observed (supplementary information Fig. 3). The flat top surfaces of the targets induce a trajectory distance difference for the droplets at the outside and inside of the cone implying the distances are no longer optimal for part of the droplets, thus generating lower efficiency. Besides, a cone forms less air wake compared to a straight droplet train so that in general the energy losses against air friction forces increase. Compared with Kelvin's induction mode, when a voltage divider was used the induction currents in the top circuits were close to the optimal value for such a system as shown before.[1] Besides, much higher currents were collected at the load resistance, enabling a higher energy conversion efficiency.

To further optimize the system we investigated a much lower gate resistance (R_g < 10GOhm) in the same symmetric connection, with the aim by tuning $R_g/(R_L+R_g)$ to create the optimal fraction of operation gating voltage over target voltage observed in our previous investigations (U_g/U_{target}=170V/20kV). [1] However, in the experiments only a slight upstream current was induced remaining constantly small within at least 10min. To explain this observation, we derived a general induction model which is presented below, where we will show that in general it is impossible to avoid either positive or negative feedback in an ohmic divider system.

Diode voltage divider

As observed in the above experiments and explained in the induction model below, an ohmic resistive voltage divider either overcharges or non-charges the droplets. To induce a stable current and consequently obtain a stable energy conversion, we therefore decided to introduce elements with a non-linear resistance behavior for the electrostatic gating in the circuit. We applied reversely connected diodes for this purpose, which exhibits an ohmic behavior at low currents and a constant voltage drop at high currents. The benefits of adding these reversely connected diodes are that the induction voltage now can become a constant independent of target voltages (or downstream current), while the induction mechanism will still work. As a result, the gate voltage can stay in a constant suitable range preventing overcharging the droplets. The I-V curve of the reversely connected diode used is shown in the Supplementary Information Figure 1. An exponentional fit to the measured diode data gave as result that the diodes have a non-ohmic resistance behavior described by U_g = 533*(1-exp(-0.55*I_i)), where I_i is the current through the diode in reversed polarity (see Supplementary information Figure 1b).

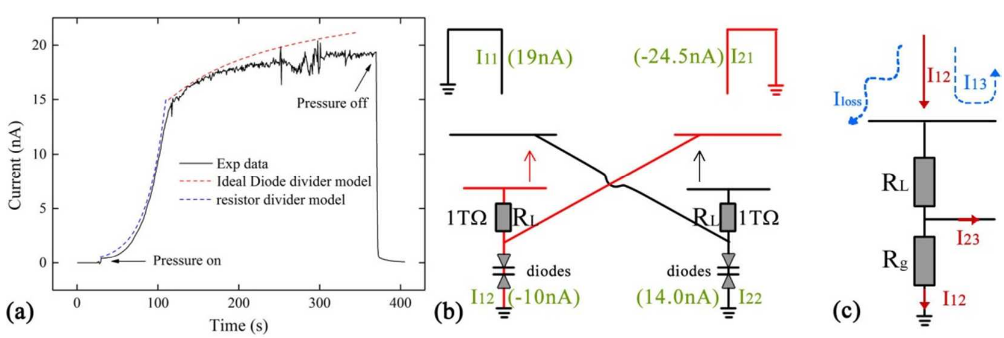

Figure 3. a. The rise of current I_{11} as a function of time when the feedback circuit with diodes is used as shown in b. A clear current saturation is observed and the current disappears immediately when the pressure is turned off. Blue dashed line shows the theretical prediction

using ohmic resistor ($R_g/(R_L+R_g)=0.02$) divider by our induction model where R_g here is the equivalent resistance of a reversely connected diode. Red dashed line shows the prediction by using reversly connected diode divider ($nC_{ind}=0.05nA/V$, $C_L=12pC$). b. Scheme of the electrical circuit and values of the measured currents. The efficiency can be calculated by simultaneous measurement of flow rate and pressure. c. the schematic picture of the current loss and gain in the circuit, taking the left jet as an example. R_g represents the resistance of diodes. The measured current are low estimate of current in downstream due to partial leak current from the (rightside) gate ring.

In the setup we furthermore added a second forward connected diode in series to prevent random charging of the two microjets (figure 3b). When the pressure was turned on, the initial current was less than 1nA and was generated by the microjet due to the sum of the streaming current and initial induction current (figure 3a). This value is lower than the streaming current measured in pure water solution (about 3 nA) because of the use of a salt solution (10mM KCl) to facilitate inductive charging decreases the membrane surface zeta potential. The current then rapidly increased when the current flow through the diodes generated an induction voltage. Above 15nA the current started to saturate, finally reaching a plateau around 19nA. Figure 3a also shows two fitted curves (dashed lines) obtained from a resistor divider model and a diode divider model (see Induction model section) respectively. It can be seen that the noise in the saturated current section is somewhat larger, but much smaller than the noise cause by deflected droplets landing on the gate. No periodic transients were observed in the current measurements within an observation period of 5 minutes, indicating that the amount of deflected droplets did not lead to water dripping from the gate. We attribute this noise to transient instability of the jet. One thing that needs to be noticed in Figure 3b is that the induction currents for the two jets are different. This can be explained by slight variations of the gate ring positions, inducing differences of the induction capacitance of the two jets; also the small differences in saturation voltage of the diodes will result in different induction voltages and hence induction currents.

It can be seen from figure 3b that a much higher downstream current than in the case of the resistor gating (figure 2) was obtained, which can be explained by the lower amount of deflected droplets. The target voltage can be approximately calculated by the sum of the voltage over the resistor and the diodes (about 500V when the downstream current is over 4nA). From the measured flow rate for the two microjets (12.3μL/s) and the applied pressure (1.40bar), we then calculate that the total efficiency of the self-excited system is at least 18%. The current flow diagram is shown in figure 3c, taking the leftside jet as an example. Current generated by jet has three destinations: landed on target, deflected and loss elswhere, while the latter two do not contribute to the energy conversion. Besides, the measured doenstream current is the low estimate since partial current flow through the rightside gate because of opposite polarity of droplets landing on the (rightside) gate ring. There is still space to improve the performance of this device, such as by changing the orifice diameter of the gate ring, optimal design of the target and optimizing the choice of the diodes. For example, the droplets partially landed on the Delrin covers (I_{loss} in Figure 3c) leading to a loss of electrical energy. A more optimal design of the complete setup will thus be useful for improving the energy conversion efficiency.

Induction model

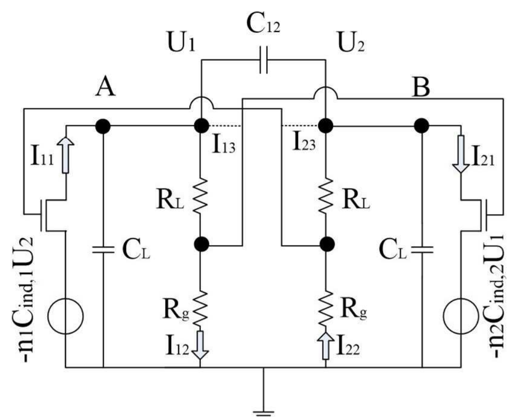

Figure 4. An equivalent circuit to model the induction mechanism in the ballistic Kelvin's water dropper. The two current sources represent the two jets 1 and 2 through which a current I_{i1} flows proportional to the gate voltage $m_j \cdot U_j$, the effective gate capacitance expressed per droplet $C_{ind,i}$ and the droplet frequency n_i. For simplicity we do not consider current loss(I_{13}, I_{23}) and charge release between two targets (C_{12} is infinitely small), the current I_i through the load plus target resistor charges the target capacitance C_L and after sufficient time the target reaches a voltage equal to $I_i(R_L+R_g)$.

An equivalent circuit to model the induction mechanism in the present device is shown in figure 4. Different from Kelvin's generator, both targets are connected to ground by resistors and voltage dividers are used for charge induction. The charge induction behavior can be considered to control the two current sources of the jets similarly as a Field Effect Transistor (FET), with a current described for jet 1 by $I_{11} = -n_1 C_{ind,1}(m \cdot U_2)$, where n_1 and $C_{ind,1}$ are the generation frequency of droplets and the effective induction capacitance of gate 1 for a single droplet, respectively, and m and U_2 are the fractional voltage division ($m=R_g/(R_L+R_g)$)

and the (opposite) target voltage, respectively. R_g is formed by either a resistor or an inversely connected diode. Here the minus sign indicates the reverse polarity of induction current and target voltage. Similarly, we can calculate the induction current on the other side as $I_{21} = -n_2 C_{ind,2}(m \cdot U_1)$.[14] To simplify the model, we don't discuss deflected droplets here, so that jet current equals current through the load.

By Kirchhoff's law, the sum of the currents flowing in and out at nodes A, B is zero. The currents at nodes A and B can be described as follows:

$$-mnC_{ind}U_2 = U_1/(R_L + R_g) + C_L\frac{dU_1}{dt} \quad (1)$$

$$-mnC_{ind}U_1 = U_2/(R_L + R_g) + C_L\frac{dU_2}{dt} \quad (2)$$

Here we assume that the single droplet current induction capacitances C_{ind} for both jets are equal. By solving these equations, we obtain the generated voltage as a function of time.

$$U_1 = \mathrm{A}\exp(\frac{nmC_{ind}}{C_L} - \frac{1}{RC_L})\mathrm{t} \quad (3)$$

$$U_2 = -\mathrm{A}\exp(\frac{nmC_{ind}}{C_L} - \frac{1}{RC_L})\mathrm{t} \quad (4)$$

Hereby A is a constant, which depends on the initial value of streaming current and gate resistance.

From equations (3) and (4), once $\frac{nmC_{ind}}{C_L} - \frac{1}{RC_L} > 0$, such as in Kelvin's water dropper (m =1, $R_L = +\infty$), a positive feedback results in the fast increases of induction current thus absolute value of the target voltage as time passes until voltage break down between two targets; while $\frac{nmC_{ind}}{C_L} - \frac{1}{RC_L} < 0$, such as considerably low resistance is used in the voltage divider, a negative feedback system results in decreasing the induction current from the initial value determined by the streaming current. Generally stated, if we use such ohmic resistance dividers that the fraction m of target voltage applied to the gate remains constant, the induction current, as seen from equations (3) and (4), will either undergo positive feedback (leading to overcharging) or negative feedback (leading to non-charging). For our system, the critical transition resistance from non-charging state to overcharging state can be calculated as $R_{L,critical} = 1/nC_{ind}$ = 20 GOhm (assuming nC_{ind} = 0.05 nA/V). Series voltage divider ratios are calculated and discussed in the supplementary information Fig 2. As shown in the above experimental results however, this resistance value led to undercharging which is not surprising as slight variations in all parameters will exist. The choice for the use of non-Ohmic reversely connected diodes is motivated by these phenomena.

Further discussion and conclusion

The experimental efficiency of the self-excited device, though reaching an appreciable value of about 18%, is much smaller than the maximal efficiency of 48% previously found when using a 30μm pore and a voltage source for gating. [1] We expect that this is mainly due to the larger working distance between gate and target in the self-excited system used here which leads to energy loss due to air friction acting on the droplets. As we showed in the supplementary information of the previous work, the system efficiency is a function of distance between target and gate ring and in the present device for ease of operation we needed to create additional space to electrically connect the gate. In future however, we expect efficiency can be improved by redesign of the gating system. Another reason for the lower efficiency, as discussed above, is the formation of a droplet spray cone instead of a droplet train due to droplet repulsion when droplets become highly charged. As a result the trajectory distances for droplets at the inside and outside of this cone are different, causing easier deflection for the droplets at the outside of the cone, decreasing the total energy conversion efficiency. It is therefore preferable to use a hemispherical target and place the pore at its center, so that all the droplets have the same trajectory distance. In addition, a better target design without Delrin cover is preferable to fully collect the charged droplets.

The self-excited ballistic energy conversion system conceptually is similar to the Kelvin's water dropper, although the systems fundamentally differ in driving force (pressure or gravity) and energy conversion mechanism (inertial to electrical energy or gravitational to electrical). Interestingly, no theoretical analysis on the energy conversion efficiency of Kelvin's water dropper has been made yet. However, theoretically the energy loss is expected to be quite low, due to the low surface energy (large droplets) and air friction (low droplet velocity). But, the real-life limitation of Kelvin's water dropper is the need for an extremely high voltage on the collector due to their large size and small the charge over mass ratio. In our case, micrometer sized droplets are produced from the pore which can have a very high charge over mass ratio, so that the target voltage can be much lower (down to hundreds of volts as shown in previous work). Besides, much less volume flow of water is needed in ballistic energy conversion than Kelvin's water dropper. Finally, it will be also possible to vary the operational current mode of the current device. A theoretical analysis and equivalent circuit were designed, to explain the unstable current induction in the resistance divider mode and stable current induction in the diode divider mode. A further refinement will be in DC or AC operation. Here we present a DC voltage microfluidic energy harvesting device. However, it has been shown that by proper design and connection of the circuit the Kelvin's droplet generator can also operate in

AC mode, which will also be true for this ballistic version. [14]

Due to the high efficiency and power density of ballistic energy conversion systems, they can become a quite competitive method for renewable clean energy. For further application in daily life this technology however needs to be developed by strongly integrating micro-pore membranes in complete closed systems, where e.g. the water transport is also managed. Systems with large pore arrays can be easily produced using cleanroom technology, so that the power can be relatively easily increased. A first application area of such devices would be in lab on a chip systems that use high voltage, such as electrokinetic separation systems. When the operational voltage of the device can be further reduced by optimizing the gate potentials to less than one kV, also other applications would become feasible.

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