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# Addendum: Self-operating transpiration-driven electrokinetic power generator with an artificial hydrological cycle

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Addendum for 'Self-operating transpiration-driven electrokinetic power generator with an artificial hydrological cycle' by Jaehyeong Bae *et al.*, *Energy Environ. Sci.*, 2020, **13**, 527–534, https://doi.org/10.1039/C9EE02616A.



In this article, we reported the extension of the operation time of the transpiration-driven electrokinetic power generator (TEPG) using calcium chloride.† Here, we aim to elucidate the potential implications for individual electrodes stemming from the intricate nature of nano-hydrovoltaic devices, a topic of recent wide-ranging discussion within this research field. Recent studies have raised concerns about electrode oxidation, potentially misattributing current generation exclusively to redox reactions. Therefore, we conducted additional experiments to evaluate the effect of redox reactions in the self-operating TEPG system. Yet, it is still difficult to specify the influence of the redox reaction on the voltage and current generation behavior of the self-operating TEPG.

In Fig. 5 of the original paper, we demonstrate that the current and voltage amplification is achieved by stacking multiple devices on a single electrode, with a linear relationship between the number of devices and amplification. If oxidation plays a dominant role, the output current would be influenced by the area of electrode participation. Since we fixed the contact area while observing the amplification, we concluded that the redox reaction is not the leading source of power generation.

Traditional electrical power generation through electrochemical reactions generally involves two separate electrodes (anode and cathode) and an electrolyte enabling ion movement. However, several factors can influence the current output in nano-hydroelectric systems where both conductor/conductor interfaces and conductor/electrolyte interfaces are present. To explore this, we performed experiments under identical conditions using various electrode materials, including Au wires, Pt wires, Ag wires, steel wires, carbon plates, and gold-plated clips, as shown in Fig. S8 (ESI†). We used wooden clips for the device and electrode connection to ensure reliable contact during the experiments. Despite minor differences, we confirmed the current generation across all electrode types.

Interestingly, even the relatively inert materials such as Au, Pt, Ag, and carbon exhibited varying current and voltage values, requiring a deeper analysis of the device–electrode interfacial contact. For instance, Liangti Qu *et al.* presented a humidity-induced nanogenerator based on graphene oxide. They reported a negligible current generation when employing an Au electrode in contrast to an Ag electrode, attributing this difference to variations in the work function at the device–electrode interface.[1,2] In our supplementary experiment, consistent with other research findings,[1,2] we observed 16 mV and 350 nA for Au electrodes, and 350 mV and 700 nA for Ag electrodes, respectively. This suggests that comprehensive analysis is needed not only for simple redox factors but also for the choice of electrode materials (such as carbon, ceramics, conductive polymer, *etc.*) considering their electronic structure and electrochemical factors at the electrode interface. This emphasizes the importance of selecting an appropriate electrode material for a device.

Furthermore, as suggested by numerous review articles, the complex interactions involving ions, electrons, and interfaces are critical for the operation of such devices.[3] In our system, interactions at the three-phase interfaces of liquid–electrode–device may unpredictably affect the surface electronic structure of each material and, consequently, impact performance. Hence, selecting

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suitable electrodes for each hydrovoltaic system is essential for overall performance enhancement. Our study demonstrates that using calcium chloride can prolong the energy generation time of the TEPG. However, we cannot definitively quantify the contribution of the electrodes' redox reactions to energy generation. If corrosion is the primary factor, it is challenging to explain the phenomenon that stacked multiple self-operating TEPGs generate amplified currents. Based on our additional experiment, we found that there are complex interactions among multi-materials and interfaces at the electrode–TEPG device junction, including the influence of contact resistance between wet carbon and electrodes with specific electronic structures. Therefore, a further comprehensive analysis of various electrodes is required to better understand these relevant mechanisms.

The Royal Society of Chemistry apologises for these errors and any consequent inconvenience to authors and readers.

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