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Static Electricity Powered Copper Oxide Nanowire Microbicidal Electroporation for Water Disinfection

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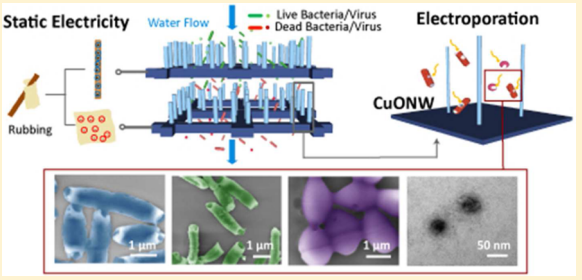
Supporting Information

ABSTRACT: Safe water scarcity occurs mostly in developing regions that also suffer from energy shortages and infrastructure deficiencies. Low-cost and energy-efficient water disinfection methods have the potential to make great impacts on people in these regions. At the present time, most water disinfection methods being promoted to households in developing countries are aqueous chemical-reaction-based or filtration-based. Incorporating nanomaterials into these existing disinfection methods could improve the performance; however, the high cost of material synthesis and recovery as well as fouling and slow treatment speed is still limiting their application. Here, we demonstrate a novel flow device that enables fast water disinfection using one-dimensional copper oxide nanowire (CuONW) assisted electroporation powered by static electricity. Electroporation relies on a strong electric field to break down microorganism membranes and only consumes a very small amount of energy. Static electricity as the power source can be generated by an individual person's motion in a facile and low-cost manner, which ensures its application anywhere in the world. The CuONWs used were synthesized through a scalable one-step air oxidation of low-cost copper mesh. With a single filtration, we achieved complete disinfection of bacteria and viruses in both raw tap and lake water with a high flow rate of 3000 L/(h·m^2), equivalent to only 1 s of contact time. Copper leaching from the nanowire mesh was minimal.



KEYWORDS: *Static Electricity, Electroporation, Nanowire, Water Disinfection, High-Efficiency*

Waterborne pathogens cause primarily enteric illnesses and remain a great threat to public health. Diarrheal illness causes more than 3 million deaths worldwide every year.[1,2] Most deaths occur in developing countries where access to clean drinking water, sanitation, and electricity is estimated to be 66%, 40%, and 21%, respectively, in sharp contrast to 99%, 99%, and 99% in developed countries.[3,4] Waterborne illness can also cause morbidity and mortality after natural disasters such as Haiti's 2010 earthquake, which caused the cholera outbreak and consequently more than 500 000 illnesses and 8231 deaths.[5] Inexpensive, fast, and effective methods of water disinfection have the potential to make a great impacts on individuals' well-being in developing countries as well as regions facing natural disasters or wars.[6,7]

Nanomaterials (nano silver, carbon nanotubes, etc.) offer new opportunities in water disinfection especially for decentralized area where commonly adopted water disinfection methods are solution reactions based or gravity-fed filtration based that rely on surface contact with the microorganisms.[8−14] Nanomaterials offer larger surface area than bulk materials and also tailorable surface active sites to improve efficiency of water disinfection[8−10,15−17] However, the large-scale deployment of nanomaterial water disinfection techniques is still challenging due to the high cost of nanomaterial synthesis and nanomaterial recovery from solution reaction system, as well as the slow treatment speed, high energy consumption, and fouling for filtration-based treatment.[18−20] We recently developed one-dimensional nanomaterial-assisted electroporation (1D-NE) to efficiently inactivate both bacteria and viruses in water with low energy consumption and without generating harmful disinfection byproducts.[21,22] 1D nanomaterials such as nanowires and nanopillars have been recently shown to interact strongly with biological cells.[23−26] In 1D-NE, the 1D nanowire structure increased the electric field by 3−4 orders of magnitude, which decreased the applied voltage to only several volts. The microorganisms were inactivated through membrane damage, which makes this method generally applicable to all type of microorganisms.[21,22,27]

We explore the use of static electricity as a power source, as this could be readily available in locations with poor access to electricity as in some regions of the developing world. Static

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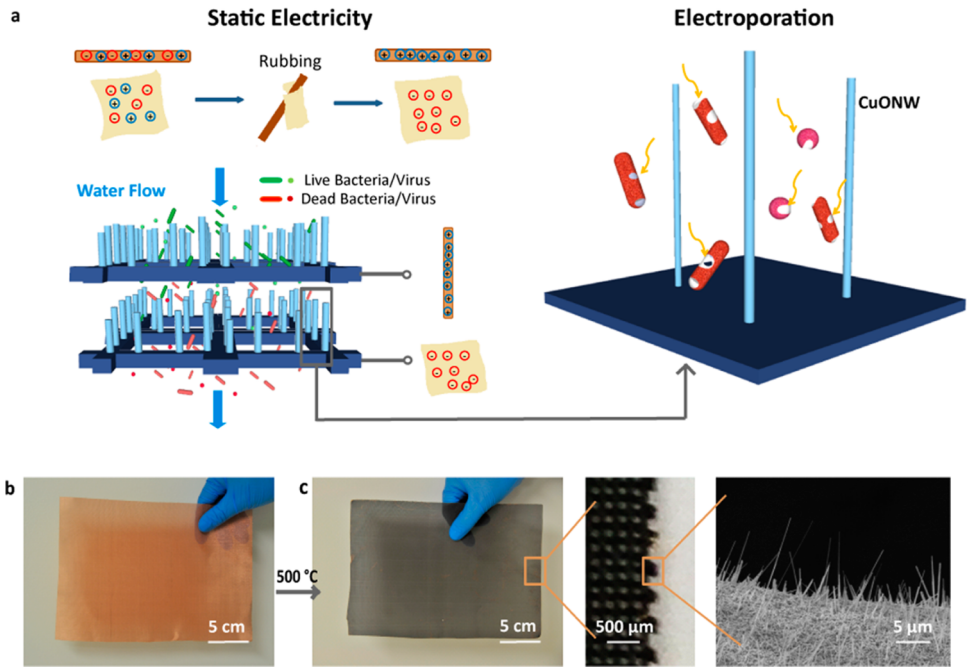

Figure 1. Schematics of static electricity powered 1D-NE water purification, CuONW synthesis, and characterization. (a) Schematics of CuONW filtration device during operation powered by static electricity. Enlarged schematics showed bacteria and viruses being electroporated at the vicinities of CuONWs. (b) Photograph of Cu mesh. (c) Photographs and SEM image of CuONW mesh synthesized from oxidizing Cu mesh at 500 °C.

charge generation was formally described in the 17th century.[28] A number of very recent studies have successfully demonstrated the use of triboelectric generators as power sources to charge portable electronics, split water, and power other electronic devices.[29−34] Human motion or other mechanical energy can be harvested to generate static electricity of tens to thousands of volts, whereas the output current is only on the order of nano amps to micro amps which is safe to people.[30,35] This unique characteristic of static electricity matches the requirement for 1D-NE, which needs a strong electric field and low current to decrease electrochemical reactions.

The configuration of the static electricity powered CuONW electroporation flow device are shown in Figure 1a. Two parallel CuONW mesh electrodes were compressed inside an in-line filter holder. During operation, static electricity was applied to the two parallel electrodes. The bacteria and viruses in the vicinity of CuONW are electroporated by the strong electric field. To reduce the complexity and cost of large-scale nanomaterial synthesis, the CuONW mesh electrode was synthesized by a simple one-step process of oxidizing Cu mesh in air at 500 °C[36] (shown in Figure 1b,c). After oxidation, the mesh became black due to the formation of CuONW. The copper mesh has a ∼150 μm pore size which is much larger than the size of bacteria (∼1−2 μm) or viruses (∼10−100 nm). The large pore size ensures a fast flow rate and avoids fouling yet such a pore size range is not too large and still ensures high probability of microorganism electroporation. A scanning electron microscope (SEM) image (Figure 1c) shows the morphology of the CuONWs. The nanowires were grown rooted to the mesh surface with diameters mostly below 100 nm and lengths above 10 μm. Therefore, direct mechanical and electrical connections of CuONWs are maintained with the Cu mesh during water flow.

The experimental setup and device photograph are shown in Figure 2a and Supporting Information Figure S1. The flow device was powered by static electricity generated by friction between a piece of plastic film and aluminum foil (see video in Supporting Information). By rubbing a piece of plastic film between two pieces of aluminum foil, voltage was generated across the foil. The foil was connected to the two electrodes in the filtration device. The voltages could be tuned by changing the contact area of the plastic film and aluminum foil. The maximum voltage of static electricity generated was measured by monitoring the voltage across a 300 MΩ resistor connected to the static electricity generating device (Figure 2b). The generated electricity showed an alternating pattern with a frequency of ∼1 Hz defined by the hand motion frequency and the peak values of 2.0 kV and −3.7 kV. This result showed that peak voltage was large enough for electroporation to occur comparing to our previous electroporation flow device which used a direct current (DC) power source with voltages from 0 to 20 V.[21]

The disinfection performance was evaluated using three model bacteria and one model virus. The model bacteria were *Escherichia coli*, *Salmonella enterica* serovar Typhimurium, and *Enterococcus faecalis*. These represent both Gram-negative and Gram-positive species, and *Salmonella enterica* serovar Typhimurium is a pathogen. The model virus was MS2, an F+ bacteriophage of *E. coli* often used as a process surrogate for human enteric viruses.[37] The inactivation performances were assessed using both raw (unaltered) tap water and raw natural lake water (Central Lake, San Mateo, California) as the aqueous phase seeded with ∼10^7 colony forming units (CFU)/mL bacteria or ∼10^7 plaque forming unit (PFU)/mL viruses. The flow rate of water through the device was maintained at 3000 L/ (h·m^2) by a peristaltic pump, which is orders of magnitude faster than the flow rate of used for filtration-based water purification (typically ∼20−65 L/ (h·m^2)).[38] The inactivation efficiencies are shown in Figure 2c−f for the different model organisms. During operation, only one pulse of static electricity was first given to the device at 60 s (Figure 2c−f) and no more pulses were supplied in order to study the

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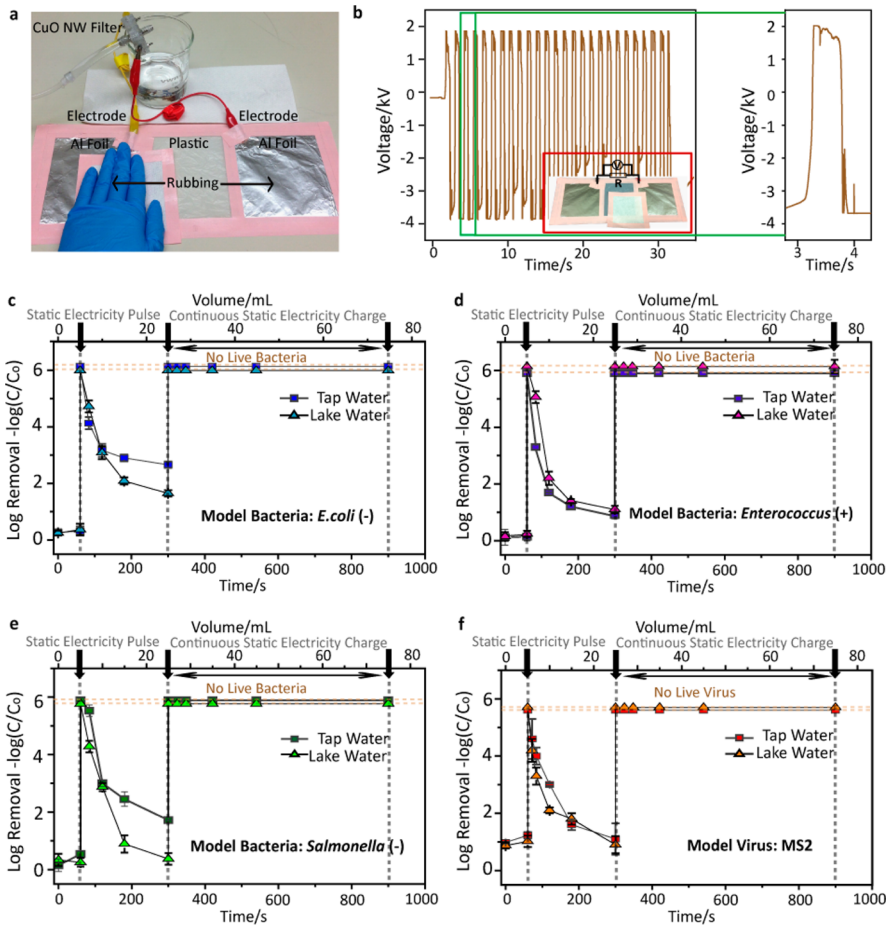

Figure 2. Experimental setup, static electricity voltage profile, and inactivation performance of model bacteria and virus. (a) Photographs of experimental setup. (b) Voltage profile of motion generated static electricity monitored on a 300 MΩ resistor. (c–e) Inactivation efficiency of both Gram-negative and Gram-positive bacteria: *Escherichia coli*, *Enterococcus faecalis*, and *Salmonella enterica* serovar Typhimurium using static electricity powered CuONW filtration device. (f) Inactivation efficiency of virus MS2 using static electricity powered CuONW filtration device. Error bars represent the standard deviation of three replicate measurements.

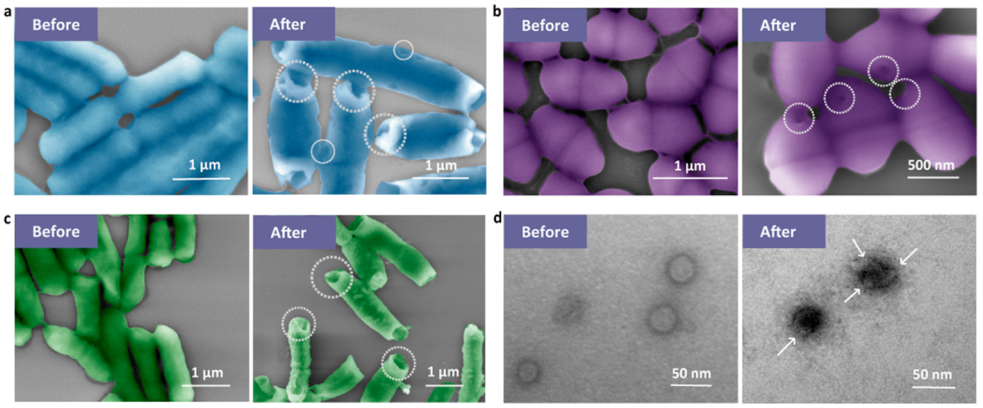

Figure 3. SEM images of bacteria and TEM images of virus before and after 1D-NE. (a–c) SEM images of bacteria before (left) and after (right) filtration by CuONW flow device. The bacteria are (a) *E. coli*, (b) *Enterococcus faecalis*, and (c) *Salmonella enterica* serovar Typhimurium. They represent both Gram-positive and Gram-negative bacteria. After filtration electroporated pores were showing formed on bacteria membranes. (d) TEM images of virus MS2 before (left) and after (right) filtration by CuONW flow device. After filtration, MS2 capsid was damaged and the inside was stained.

disinfection effect of a single pulse. Water that passed through the device during the pulse was shown to contain no cultivatable organisms. It is remarkable that even 30 s after the single pulse, the disinfection efficiency remains at a high value of >99.9% for all four model microorganisms bacterial and viral removal decayed with time owing to the decay of the electric static charge although the decay rate is slow. We define a parameter to quantify the decay of microbial disinfection, a half time $t_{1/2}$. A half time, $t_{1/2}$, is the time when the log removal decreases to half of the maximum value. For the four model

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microorganisms *Escherichia coli*, *Salmonella enterica* serovar Typhimurium, *Enterococcus faecalis*, and MS2, the $t_{1/2}$ is ~120, 60, 30, and 65 s for tap water condition and ~65, 50, 45, and 40 s for lake water condition. Bacteria and virus seeded into lake water sample (conductivity of 2.3×10^{-2} S/cm) showed a relatively faster decay rate than bacteria and viruses seeded into tap water (2.0×10^{-5} S/cm), which suggests that the static charge on the CuONW mesh electrodes surface leaks faster in solution with higher conductivity. The remarkable disinfection effect of even a single pulse suggests that continuous pulses with adequate frequency should be able to disinfect continuously with high efficiency. After 300 s, the device was powered by continuous charging of static electricity generated by continuous movement at ~1 Hz. Both the filter-sterilized tap water and lake water showed at least 6 log (99.9999%) disinfection of bacteria and 5.7 log (99.9998%) removal of virus MS2, which is no cultivatable bacteria or viruses detected (i.e., all measurements were below our detection limit so greater disinfection was not measurable).

Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) were used to characterize bacteria (Figure 3a–c) and viruses (Figure 3d and Supporting Information Figure S3) before and after filtration. The SEM images showed that after filtration, bacterial membranes were damaged and pores were formed on the cell surfaces suggesting the electroporation inactivation mechanism. For the Gram-negative bacteria *E. coli* and *Salmonella enterica* serovar Typhimurium, large pores (>500 nm) were formed and they were mainly observed on the two ends of the rod shaped bacteria, whereas some small pores were also observed on the sides. For the Gram-positive bacterium *Enterococcus faecalis*, which appeared oval in the SEM image, the pores formed on the bacteria surfaces were also mostly at the prolate base of the oval but of smaller sizes (~100 nm). The differences in pore sizes may be due to both cell surface membrane properties and cell size differences between *Enterococcus faecalis* and the other two bacteria. Gram-positive bacteria have a thicker layer of peptidoglycan than Gram-negative bacteria, which makes them more resistible to physical forces.[39] *Enterococcus* is smaller than the other tested bacteria, and smaller bacteria require stronger electric field to achieve the same extent of electroporation as bacteria of larger sizes.[40,41] TEM was used to characterize MS2 before and after treatment due to their small size (<50 nm). A negative staining method was used to investigate whether the capsid integrity of MS2 was maintained.[42,43] If the capsid is damaged, the inside of MS2 will be stained and show a dark contrast. The TEM images showed that in the filter-treated sample, the MS2 was stained dark, which indicates capsids were damaged. In addition, in the TEM image of treated MS2, some parts of the MS2 capsid were missing. This suggests electroporation causes capsid damage and is responsible for viral inactivation.

To demonstrate the importance of the nanowire structure in enhancing the electric field, the inactivation performances were compared between filters made of CuONWs and copper oxide nanoparticles (CuO_xNP) using *E. coli*. The CuO_xNP mesh electrodes were fabricated by oxidizing copper mesh at lower temperature 200 °C for 4 h. Instead of forming nanowire structures as occurs at higher oxidizing temperature, nanoparticles were formed uniformly on the surface of copper mesh (Figure 4a, b). Inactivation was first evaluated using a static electric pulse. Both CuONW and CuO_xNP showed similar decay patterns. However, the inactivation performance of

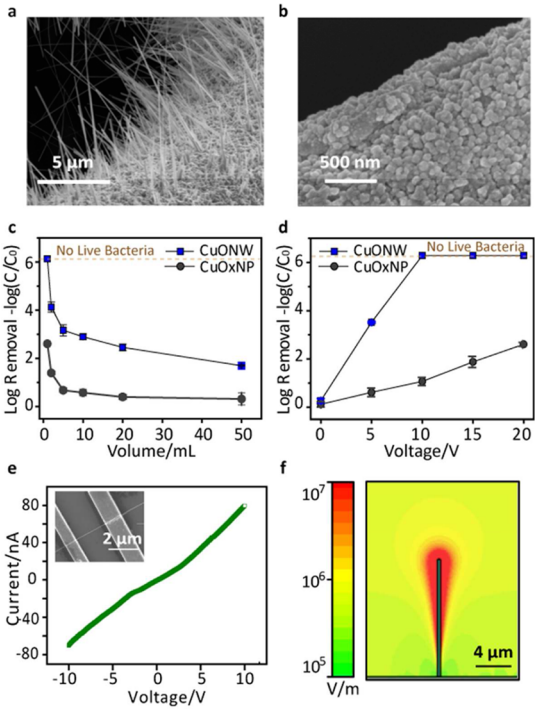

Figure 4. CuONW electric field enhancement performance and simulation. SEM images showing (a) the CuONW oxidized at 500 °C and (b) CuO_xNP oxidized at 200 °C. (c) Inactivation efficiencies by static electric pulse for *E. coli* using CuONWs and Cu_xONPs showing enhanced performance by 1D nanowire structure. (d) Inactivation efficiencies by DC power of 0–20 V for *E. coli* (e) Conductivity measurement for a single CuONW. Inset showing the SEM image of the single nanowire device. (f) Electric field distribution at 10 V of power near the surface of CuONW (diameter, 100 nm; length, 15 μm) in water showing the enhancement of the electric field strength. Error bars represent the standard deviation of three replicate measurements.

CuONW filter was at least two orders of magnitude better than CuO_xNP filter (Figure 4c). This is because after a given pulse, the voltage between the two electrodes was moderate or low most of the time, due to the decay characteristic of the electrode charge. At these moderate or low voltages, the CuONW electrode, which could locally enhance the electric field strength, has higher inactivation efficiency. The inactivation of *E. coli* achieved by the CuONW and CuO_xNP filters at voltages between 0 and 20 V supplied by a voltage source meter is shown in Figure 4d. Live *E. coli* was not detected in filtrate when using CuONW mesh electrodes at applied voltages above 10 V. Inactivation was greater when the filter was composed of the nanowire structure than nanoparticles, suggesting that nanowire morphology is better than nanoparticle morphology in enhancing the electric field. The sharp tip structure of nanowire can concentrate the electric field in the vicinity to a greater extent than nanoparticles, hence yielding a greater inactivation efficiency.

The conductivity of CuONW is essential in building the electric field and the nanowires should have higher conductivity than the water medium in order to form a locally enhanced electric field. The conductivity of CuONW was determined by a single nanowire electrical transport measurement. The CuONWs had conductivities ranging from ~10^2 S/cm (Figure 4e). Comparing to city water 5×10^{-5} S/cm and seawater 5×10^{-2} S/cm, CuONWs are conductive enough to build a strong electric field in wide range of natural waters. Electroporation,

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when used for transforming microorganism in molecular cloning, requires the electric field to be larger than 10^6 V/m.[40,44] The electric field simulation in Figure 4f showed that at an applied voltage of 10 V, the electric field at the vicinity of CuONW was above 10^7 V/m. This is high enough to generate electroporation to bacteria and viruses.[21,22,40,44] In comparison, an electrode of same material but with a flat surface under the same condition would only produce $\sim 10^4$ V/m field.

From the application prospective, the CuONW filtration device can also be powered by DC, alternating current (AC) electricity source, or battery. The disinfection performance using different power sources was shown in Supporting Information Figures S4 and S5. Copper leaching (Supporting Information Figure S6) was evaluated for all operation condition and the Cu concentration in the effluent was shown to be minimal and much lower than drinking water standard.

Methods. *CuONW Filtration Device Fabrication.* Copper mesh (McMaster, disinfection efficiency test using 100 × 100 mesh size) was etched with 1 M hydrochloric acid to remove the oxide layer and then oxidized in air at 500 °C. The CuONW mesh was cut into 1 cm × 1 cm square shape electrodes and put into in-line filter holder with a Kimwipe paper in between as a separator.

Bacteria and Viruses Inactivation. Bacteria, *Escherichia coli* (JM109, Promega), *Salmonella enterica* serovar Typhimurium LT2 (ATCC 700720), and *Enterococcus faecalis* (ATCC 19433), were cultured to log phase, harvested by centrifugation at 900*g*, washed twice with DI water, and suspended in raw tap water or filtered lake water to $\sim 10^7$ CFU/mL. Bacterial concentrations in the influent and effluent were measured using standard spread plating techniques. Each sample was serially diluted and each dilution was plated in triplicate and incubated at 37 °C for 18 h. Viruses, bacteriophage MS2, were grown with the *E. coli* Famp host on a shaker table set to 25 rpm at 37 °C for 24 h. MS2 was isolated and concentrated using the polyethylene glycol (PEG) precipitation method.[45] A solution of $\sim 10^7$ PFU/mL was made using raw tap water or filtered lake water. MS2 was enumerated using a double agar layer method. All plating was done within 3 h of filtration experiment. Influent and effluent concentrations were compared to determine the extent of inactivation.

Bacteria and Virus Sample Preparation for SEM and TEM. All bacteria samples were pelleted by centrifuging at 900*g* for 10 min and the supernatant was removed. All samples were fixed using a solution of 0.1 M sodium cacodylate buffer (pH 7.3), 2% glutaraldehyde, and 4% paraformaldehyde and stored at 4 °C overnight. Then the cells were washed with the same buffer for 5 min. Samples were dispersed on silicon wafers and air-dried in preparation of SEM characterization. A total of 20 μL of the virus samples were pipetted on a TEM grid and let the samples sit for 15 min. Samples were stained with 2% uranyl acetate solution for 45 s, and then the stain was removed.[42,43] The TEM grids were air-dried for TEM characterization.

CuO Nanowire Conductivity Measurement. The conductivity of the CuONW was determined using a single nanowire device. The CuONW mesh was sonicated in water to form a suspension containing individual nanowires. The nanowires were dropcast onto an oxidized silicon substrate and patterned into devices by means of standard electron-beam photolithography and thermal evaporation of chromium/gold (10 nm/190 nm) contacts. The nanowire device was then measured using an Agilent B1500A semiconductor device analyzer.

Electric Field Simulation. The electric field simulations were conducted using the commercial software package CST EM Studio. The CuONW was attached to one of the two flat electrodes. Pores are perforated on the electrodes mimicking the experimental environment. The distance and voltage between the two electrodes were 200 μm and 10 V, respectively.

ICP-MS Measurement. All samples were treated by adding nitric acid to 5%. The samples were filtered with 0.2 μm pore size filters before measurement.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Details about device configuration, more TEM characterizations on MS2, device performances using DC and AC power source and ICP test, respectively. This material is available free of charge via the Internet at http://pubs.acs.org.

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Author Contributions

C.L. and Y.C. conceived the concept. C.L. conducted most of experiments and X.X., W.Z., J.Y., D.K. contributed parts of experiments. C.L., A.B.B., and Y.C. analyzed the data and cowrote the paper. All the authors discussed the whole paper.

Notes

The authors declare no competing financial interest.

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