Flexible Pseudocapacitive Electrochromics via Inkjet Printing of Additive-Free Tungsten Oxide Nanocrystal Ink

Long Zhang, Dongliang Chao, Peihua Yang,* Louis Weber, Jia Li, Tobias Kraus,* and Hong Jin Fan*

Direct inkjet printing of functional inks is an emerging and promising technique for the fabrication of electrochemical energy storage devices. Electrochromic energy devices combine electrochromic and energy storage functions, providing a rising and burgeoning technology for next-generation intelligent power sources. However, printing such devices has, in the past, required additives or other second phase materials in order to create inks with suitable rheological properties, which can lower printed device performance. Here, tungsten oxide nanocrystal inks are formulated without any additives for the printing of high-quality tungsten oxide thin films. This allows the assembly of novel electrochromic pseudocapacitive zinc-ion devices, which exhibit a relatively high capacity (≈260 C g\(^{-1}\) at 1 A g\(^{-1}\)) with good cycling stability, a high coloration efficiency, and fast switching response. These results validate the promising features of inkjet-printed electrochromic zinc-ion energy storage devices in a wide range of applications in flexible electronic devices, energy-saving buildings, and intelligent systems.

The increasing demand for multi-functional flexible systems has led to the development of metal ion intercalation based electrochemical devices\(^{[1-5]}\) (e.g., metal-ion batteries, supercapacitors, and electrochromic devices) that are suitable as integratable power source for future smart devices. In particular, there has been a surge of interests in electrochromic devices because of their tunable optical transmittance, low power consumption, and their wide potential applications in smart windows, flexible displays, military camouflage, sensors, etc.\(^{[6,7]}\) Tungsten oxide (WO\(_x\)) based materials are deemed the most promising candidate as the cathode option of visible light modulated electrochromic devices, because of their high optical contrast ratios and superior cycling stability.\(^{[8-10]}\) In addition, comparing with monovalent ions, low-cost multivalent-cation electrolytes (e.g., Zn\(^{2+}\), Mg\(^{2+}\), or Al\(^{3+}\))\(^{[11,12]}\) which can provide multiple charges during redox reactions, are considered to be attractive candidates for electrochromic functions.

With the growing expectation for individual patterned design of electrochromics, traditional physical evaporation and spin coating preparative methods can no longer meet the requirements.\(^{[13]}\) Alternately, inkjet printing is a versatility technology and compatible with large-scale digital printing of customized patterns, which also has the potential advantage of reduction in material waste.\(^{[14-19]}\) It is in principle a suitable technology for printing thin films for the above mentioned electrochemical devices with more features such as flexibility and high integration. However, the inkjet printing technology needs further amelioration to overcome some obstacles, such as nozzle clogging issues and development of stable inks to print devices effectively.\(^{[20,21]}\) To date, no standard protocol has been defined to make suitable inks for inkjet printing. This is an area of great potential but not yet fully explored. In most printable inks including the WO\(_{x}\) for electrochromics, additives (such as surfactants and secondary solvents) are usually needed to tune the rheological properties of the ink,\(^{[22]}\) and to achieve uniformity of the printed patterns. The additives need to be removed by high-temperature annealing, which not only introduce synthesis complexity, but also may deteriorate most of flexible substrates. Hence, it is vital to develop new formula for high-quality inks, preferably involving no additives during synthesis towards a low-cost and reliable inkjet printing of films.

Herein we intend to develop high-quality inkjet-printed WO\(_{3-x}\) films and construct electrochromic energy storage devices. We report the synthesis of concentrated additive-free nanocrystal WO\(_{3-x}\) colloidal inks via a facile wet-chemical route. The ink was used to inkjet print electrochromic devices...
with high printing efficiency and uniformity. The devices exhibited good electrochromic performance including high coloration efficiency ($57.4$–$97.7 \text{ cm}^2 \text{ C}^{-1}$ at the wavelength of 633 nm), rapid switching response (coloration/bleaching time of 4.5/3.7 s under 0/1 V voltage profile), and good mechanical flexibility. As a multi-functional device, the as-fabricated electrochromic device can also serve as a power supply, due to a high-rate pseudocapacitive Zn ion intercalation process. This work highlights the potency of WO$_{3-x}$ nanocrystal inks for simulation for inkjet printing of electrochromic energy devices and quantifying their system-level performance, which may find applications for future flexible electronics and smart windows.

Figure 1a illustrates the synthesis of WO$_{3-x}$ nanocrystals as detailed in the experimental section. Transmission electron microscopy (TEM) and X-ray diffraction (XRD) were used to determine the morphology and the crystalline phase of as-synthesized WO$_{3-x}$ nanocrystals. XRD patterns were analyzed by a full pattern fitting technique using the general structure analysis system (GSAS) with the EXPGUI interface, confirming that the as-synthesized WO$_{3-x}$ comprises cubic unit cell with the space group of $Pm\bar{3}m$. The lattice parameter was calculated to be $a = 0.354$ nm with Rietveld refinement ($\chi^2 = 1.941$, $R_w = 7.97\%$, $R_p = 6.35\%$). The $d$-spacing of 0.36 nm indexed in the TEM image can be assigned to the (100) plane spacing. The size distribution of the WO$_{3-x}$ nanocrystals is in the range of 3–5 nm in diameter, as shown in Figure 1b.

Ink formulation was performed using the inverse Ohnesorge number $Z$, defined as the ratio of the Reynolds number $\left(Re = \frac{\rho d^4}{\eta}\right)$ and the square root of the Weber number $\left(We = \frac{\rho v^2 d}{\gamma}\right)$:

$$Z = \frac{Re}{\sqrt{We}} = \frac{\sqrt{\rho d^4}}{\eta}$$

A value of $Z$ in the range of 1–10 is required for the formation of stable and separated droplets. Note that $Z$ only depends on ink surface tension ($\gamma$), density ($\rho$), viscosity ($\eta$), and nozzle diameter ($d$), but not on the droplet velocity ($v$). We found that inks based on ethanol (EtOH), isopropanol (IPA), and N-Methyl-2-pyrrolidone (NMP) provide good droplets for inkjet printing even without any additives (Figure 1e, Figure S1 and Table S1, Supporting Information). We chose NMP for the ink used here because its boiling point over 200 °C prevents unwanted evaporation and clogging of nozzles. The resulting ink is more stable than previously reported materials and still could work effectively after one month (Figure S2, Supporting Information).

The velocity of the droplets depends on the waveform used to drive the piezo print head (Figure S3, Supporting Information). We optimized it in order to obtain droplets of uniform size and prevent satellites. Figure 1f shows a stroboscopic image of 10 pL droplets during one jetting cycle. The velocity of the droplet...
suitable for uniform printing. Figure 1e, corresponding to a Z value of ca. 1.1, that is believed to the number of printed layers. The optical transmission relative to the bare substrate first decreased with thickness due to interference-based antireflective properties and then increased as expected.[27,28] Microscale analysis via scanning electron microscopy (SEM) and atomic force microscopy (AFM) indicated a copy (SEM) and atomic force microscopy (AFM) indicated a relationship in Figure 2f confirms it is ohmic behavior, I−V plot in Figure S4, Supporting Information).[29] The linear I−V relationship in Figure 2f confirms it is ohmic behavior, and the value of conductivity calculated (=3.4 × 10^-5 S cm^-1) is slightly above reported for pure WO3 (=10^-6 S cm^-1) as expected due to the existence of oxygen vacancies.[30, 31] It is noteworthy that, while the thickness of printed thin films increases, the electrical conductivity of the films with different printing layers remain nearly the same (Figure 2g), indicative of a reliable printing quality.

Printed WO3−x films were combined with zinc foil frames as counter electrodes in order to assemble optically controllable electrochromic energy devices (Figure 3a). Figure 3b describes the cyclic voltammetry (CV) curves of the as-prepared devices with different printing layers, at a scan rate of 10 mV s^-1. The anodic peak currents derived from CV exhibits a linear relationship with the number of printed layers (shown in Figure S5, Supporting Information), which indicates that the capacity varies in proportion to the number of printed layers. It provides an additional evidence to the reliable printing quality. Considering the electrochemical performance and the optical characteristics of the printed WO3−x film, we chose the <N> = 2 sample for the following study. From the charge–discharge curves depicted in Figure 3c, we obtain nearly reversible capacities of approximately 260, 240, 200, and 180 C g^-1 at the current densities of 1, 2, 4, and 8 A g^-1, respectively. Previous literature indicates the areal performance metrics are also important for flexible devices.[32,33] Taking the current density of 2.4 mA cm^-2 (8 A g^-1) for example, the device areal capacitance is 54 mF cm^-2, which is comparable to the sputtered WO3 film (72 mF cm^-2 at 5 mA cm^-2).[30] The areal capacitance can be improved by increasing printing layers. The excellent retention of the capacity during charging and discharging (i.e., high Coulombic efficiency) indicates that the charge storage process is reversible, and the device is stable.

In the following, we analyze the optical properties of the device. Transmittance measurements at different potentials (Figure 3d) indicate an electrically modulated optical density in a wide wavelength range from 400 to 800 nm. The maximal optical modulation calculated at 633 nm was 76%, which is higher than the NiO/WO3 electrochromic battery (35%)[34] and tungsten molybdenum oxide Zn-ion electrochromic batteries (62%)[35] under comparable conditions. The intense light absorption can be assigned to the reduction of W^6+ ion when Zn ions insert into the WO3−x cathode.[35,36]

Coloration efficiency (CE) is an effective criterion for the evaluation of electrochromism defined as the change in optical density (ΔOD) of the printed film in its colored (optical transmittance Tc) and bleached (Tb) state evoked by a

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**Figure 2.** Physical characterization of inkjet-printed WO3−x films. a) Photographs of WO3−x films with different printing layers, denoted by <N>. b) Optical transmittance spectra with different printing layers. c) SEM images, and d) AFM images of the <N> = 1 sample. e) Height profiles of the edge of the printed films. f) I−V curves of the film printed on bare PET substrate. g) Film thickness and conductivity of the films.

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ranged between 5 and 10 m s^-1, as shown in the Re-We plot in Figure 1e, corresponding to a Z value of ca. 1.1, that is believed suitable for uniform printing.

We used the nanocrystal ink to print uniform WO3−x films on ITO-PET substrates (Figure 2). Film thickness was readily adjusted between 50 and 200 nm by changing the number of printing layers. The optical transmission relative to the bare substrate first decreased with thickness due to interference-based antireflective properties and then increased as expected.[27,28]
charge density \( Q \), charge intercalated into cathode materials per unit area):

\[
CE = \frac{\Delta OD(\lambda)}{Q}
\]

(2)

\[
\Delta OD(\lambda) = \log \frac{T_0}{T_{\lambda}}
\]

(3)

Our device has CE values at a wavelength of 633 nm that ranged between 57.4 and 97.7 cm\(^2\) C\(^{-1}\) depending on potential (Figure 3e). Figure 3f demonstrates the reversibility of the electrochromic switching behavior between an applied potential of 1 and 0 V. The switching times of bleaching and coloring were 3.7 and 4.5 s, respectively, corresponding to a 90% transmittance change.\(^{[13]}\) Our printed device shows a faster bleaching and coloring response compared to nanosheet WO\(_3\) film electrochromics (7.0 s for bleaching and 10.7 s for coloring).\(^{[37]}\) Inkjet printing enables rapid patterning of the electrochromic film. Figure 3g shows a digitally printed pattern that rapidly changed color when changing the potential from 0 to 1 V.

Several inkjet-printed electrochromic devices are summarized in Table S2, Supporting Information to compare the present work with state-of-art.\(^{[38–49]}\) Our WO\(_{3-x}\) nanocrystals show a much smaller size, and the formulated ink concentration is quite high, which is beneficial for uniform printing. In addition, the assembled WO\(_{3-x}\) zinc-ion electrochromic energy device also provides high coloration efficiency and compatible working voltage window.

To elucidate the electrochemical charge storage mechanism, sweep-rate dependent CV experiments were performed in order to quantitatively analyze capacitive effects in the ion storage process. Figure 4a shows the CV curves of the device with \(<N> = 2\) WO\(_{3-x}\) film at different scan rates varying from 10 to 50 mV s\(^{-1}\). We chose relatively high scan rates since their CV curves indicate strong capacitive behavior. It is noted that the CV curves in Figure 4a maintained a similar shape with evident shift of the anodic peak with increasing scan rate. The capacitive effect could be qualitatively derived by the fitting the relation between the peak current (\(i\)) and the scan rate (\(\nu\)).\(^{[50,51]}\) As this has been a standard practice in the electrochemical energy storage community, we will not repeat the details here but just present the result. In our system, the linear \(\log i - \log \nu\) plot has a gradient of 0.98 (Figure 4b), which indicates that the Zn-ion reaction is
dominated by capacitive behavior (i.e., intercalation-type pseudocapacitance). Further fitting of the CV curves at different scan rates can provide quantitative information to distinguish capacitive contribution from diffusion-controlled contribution to the total capacity. Figure 4c depicts a voltammetric response at a scan rate of 30 mV s⁻¹ of the capacitive contribution compared with the total measured current. By comparing total energy storage and that contributed from capacitive process, we found that at a slow scan rate of 5 mV s⁻¹, the capacitive effect contributes more than 70% of the total energy, and the value increases to 97% at a higher scan rate of 100 mV s⁻¹. This analysis confirms that energy storage in the inkjet-printed WO₃₋ₓ film exhibits a capacitive dominating behavior, accounting for its fast charge/discharge response. Elezzabi et al. proposed that zinc ions intercalation into tungsten oxide is the key to triggering electrochromism.⁵⁻⁶ Considering the high capacity and the fast response time of our electrochromic energy device (Figure 3f), it is reasonable to infer that an intercalation pseudocapacitive behavior predominates in our system.

To verify the Zn ions intercalation, we replaced the ZnSO₄ electrolyte with H₂SO₄ that has a similar pH value. As shown in Figure S6, Supporting Information, the capacity decreases significantly with the H₂SO₄ electrolyte. This implies that the proton has a limited effect on the electrochemical properties and negligible capacity contribution. Therefore, herein we consider zinc ions are the main charge species.

To further understand the diffusion kinetics of zinc ions, we measured the AC impedance of the samples with different applied potential, at frequencies from 200 kHz to 0.1 Hz (Figure 4e). We propose that the overall model consists of three parts in series: i) the series resistance of the testing configuration (R₀), ii) the resistance of ions diffusing within the electrochromic film (R₁), capacitive behavior of ion diffusion in the solid (CPE₁), and iii) the resistance of charge transfer across the electrolyte/electrode interface (R₂), Warburg impedance (W) related to ions diffusing at the electrolyte/electrode interface and rapid diffusion at the near-surface region,⁵¹ and the relative double-layer capacitance (CPE₂).⁵²⁻⁵⁶ In the region of high frequency, the change in resistance R₂ is insignificant, and it is independent with the applied potential (Figure S7, Supporting Information). However, for R₁ and the Warburg diffusion element, they change as a function of the applied voltage and reach the minimum value at 0 V. So, it can be inferred that a gradual transition from diffusion-controlled process to surface charge transfer-controlled process will take place as the zinc ions reach to the WO₃₋ₓ film. This is in good agreement with the impedance of the Li⁺-WO₃ system, as studied earlier by Ho and coworkers.⁵² To determine the zinc ions diffusion coefficient in the WO₃₋ₓ electrode, potentiostatic intermittent titration technique (PITT) was carried out, as demonstrated in Figure S8, Supporting Information. The resulted diffusion coefficient for zinc ion is in the order of 10⁻¹² cm² s⁻¹, which is comparable to the lithium ions diffusion coefficient in tungsten oxide electrodes.⁵³

We tested the performance of the overall device to better judge its suitability for possible applications. Figure 5a shows
CV curves at 50 mV s\(^{-1}\) of devices that were bent at 0°, 90°, and 180° and found little effect on the performance. In addition, the capacity remains 80% after 100 times 90° bending measurements (Figure S9, Supporting Information), indicating good mechanical flexibility. Figure 5b indicates that over 70% of the initial capacity is retained after 1000 charge/discharge cycles. The capacity loss during cycling is mainly attributed to film cracks and materials exfoliation, which is common in thin-film based energy storage devices. Four series-wound devices can light up a red light-emitting diode (LED) directly, as shown in Figure 5c. After 5 min, the LED light almost fades out, and the four cells all turned to a light blue state. This means that we can estimate the energy storage/consumption condition directly by visual sight and for possible application in smart windows.

We have developed a high-quality nanocrystal-based ink that is suitable for inkjet printing functional electrochromic devices with reliable printing quality. Concentrated nanocrystal colloidal inks without any additives are stable up to one month, which can effectively reduce the material waste and lower the printing cost. The assembled electrochromic devices show both excellent rechargeable capacitive energy and electrochromic performance, including high coloration efficiency, fast switching response, good cycle stability, as well as flexibility. The reversible energy storage synchronized with optical modulation makes this electrochromic device potentially an intelligent power source. The ink formulation strategy and printing technique provide new possibilities for applications in smart flexible electronics, sensors, and energy-saving fields.

**Experimental Section**

**Materials:** Indium tin oxide coated polyethylene terephthalate (ITO-PET, thickness 0.13 mm, thickness of ITO coating is 1300 Å, 60 Ω sq\(^{-1}\), transmittance > 78% @ 550 nm), tungsten (VI) chloride (WCl\(_6\), 99.9\%+), oleylamine (tech. grade 70%), triethyloxonium tetrafluoroborate (97.0\%), N,N-Dimethylformamide (DMF, 99.8\%), N-Methyl-2-pyrrolidinone (NMP, 99.5\%), hexane (97.0\%), acetonitrile (99\%), zinc sulfate heptahydrate (99.0\%), and zinc foil (99.9\%) were purchased from Sigma-Aldrich; Oleic acid (tech. grade 90\%) was from Alfa Aesar.

WO\(_{3-x}\) Nanocrystals Synthesis and Ink Formulation: The synthesis method was modified from D. J. Milliron’s previous report.\[^2\] Briefly, 20 mL oleic acid (OA) was mixed with 2 mL oleylamine (OAm) and degassed under vacuum at 120 °C for 1 h, and then transferred to an oil bath at 300 °C with Argon flow. Then, 400 mg WCl\(_6\) powder was added to 4 mL oleic acid under stirring, and the precursor then was injected into the OA and OAm solvent mixture. The solution quickly turned to a dark blue color and was kept for 5 min. When the solution was cooled down to room temperature, 44 mL isopropanol (IPA) and 10 mL deionized (DI) water were added. The nanoparticles were centrifuged and washed twice by IPA, and finally dispersed in 10 mL hexane. The organic surface coordinating ligands were stripped by the following method: 500 mg triethyloxonium tetrafluoroborate was dissolved in 50 mL of acetonitrile, mixed with WO\(_{3-x}\) dispersion in a Schott bottle, which was then stirred overnight under ambient condition. The nanocrystals remained stable during storage in the ligand exchange solution and could be dispersed in polar solvents. Before the inkjet printing process, the precipitated stripped nanocrystals were redispersed into 2 mL NMP to

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**Figure 5.** Demonstration of the electrochromic energy device. a) Flexibility test by measuring the CV curves at 50 mV s\(^{-1}\) under various bending conditions (photographs shown as inset). b) Long-term cycling performance at 20 mV s\(^{-1}\) for 1000 cycles. The inset shows the CV curves at the 1st and 1000th cycle. c) A light-emitting diode (LED) is powered by four series-connected electrochromic cells. Note that the electrochromic cell becomes opaque when the LED light dims.
formulate WO$_3$ nanocrystals ink. The final concentration of WO$_3$ was approximately 100 mg mL$^{-1}$.

**Inkjet Printing Process:** The WO$_3$ ink was filtered through a 0.2 µm filter before printing and subsequently printed using a PiXdro LP50 printer equipped with a 21.5 µm diameter inkjet nozzle. Before printing, the ITO-PET substrate was activated by oxygen plasma for 20 min; the substrate was maintained at 60 °C during the printing process. The printed WO$_3$ film was annealed at 120 °C for 30 min before the following measurements. The mass loading of each printing pass is about 0.15 mg cm$^{-2}$.

**Characterizations:** Scanning electron microscopy (SEM) imaging was conducted on a Quanta 400 SEM (FEI, Germany). Transmission electron microscopy (TEM) image was recorded using a NanoWizard3 machine (JPK Instruments, Germany). The surface tension was measured by DataPhysics Instrument GmbH (OCA 35). The viscosity of the inks was measured by Discovery Hybrid Rheometer (HR-3). Keithley 2450 electrometer was employed to derive conductivity using the four-probe method. In situ transmittance spectra of the WO$_3$ film were performed using a UV–vis spectrophotometer ( Cary 5000). Electrochemical measurements were proceeded on an electrochemical workstation (VMP2, Biologic) using 1 mol L$^{-1}$ ZnSO$_4$ electrolyte.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Keywords**

additive-free inks, flexible electrochromics, inkjet printing, zinc-ion storage, tungsten oxide

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