**Supporting Information**

Combustion Synthesis and Photoelectrochemical Characterization of Gallium Zinc Oxynitrides

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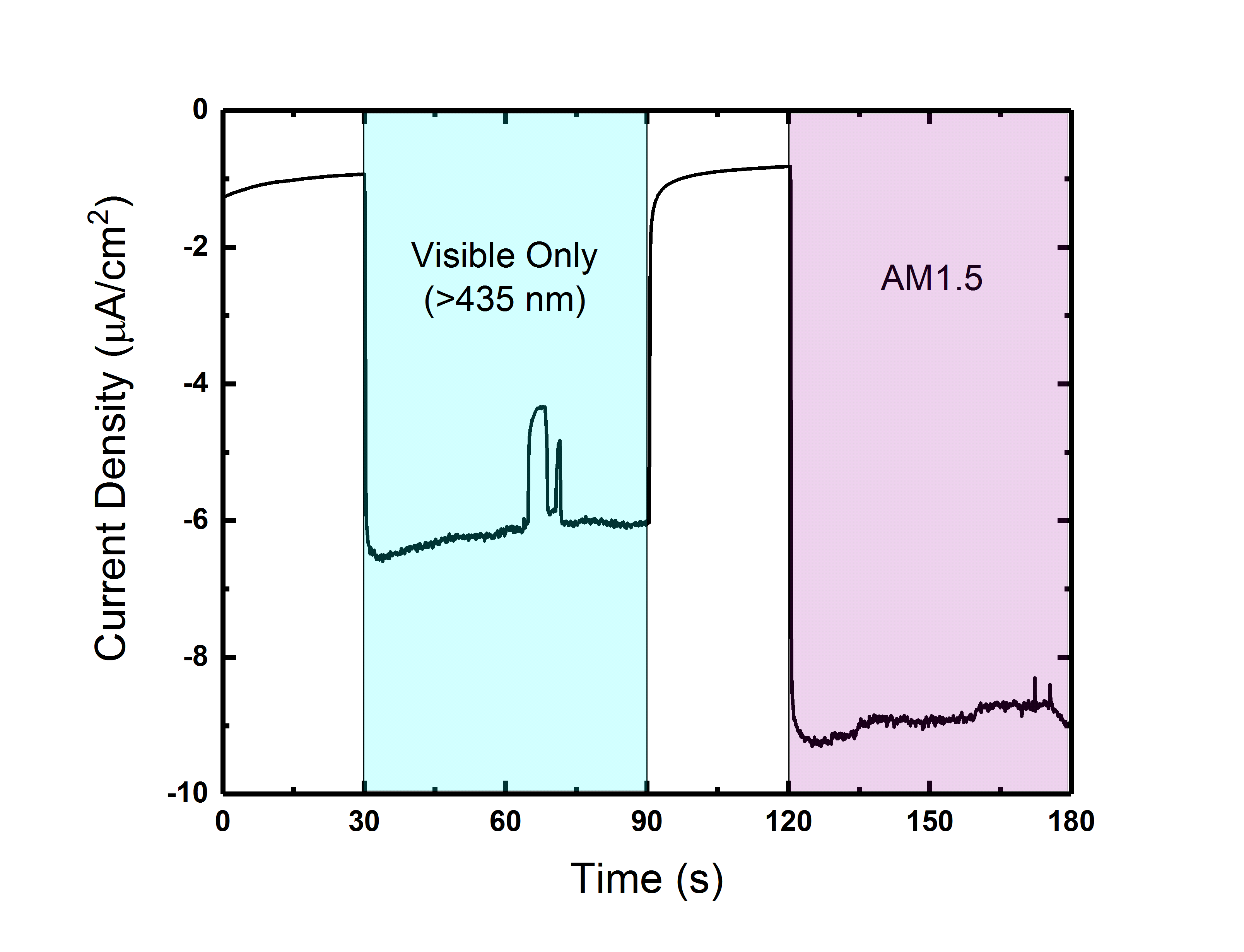
**Figure S1**: A) Ga 2p and B) Ga 3d XPS spectra of the 1:1:10 GaxZn1-xOyN1-y sample. Peaks were assigned according to literature and confirm the presence of GaN.



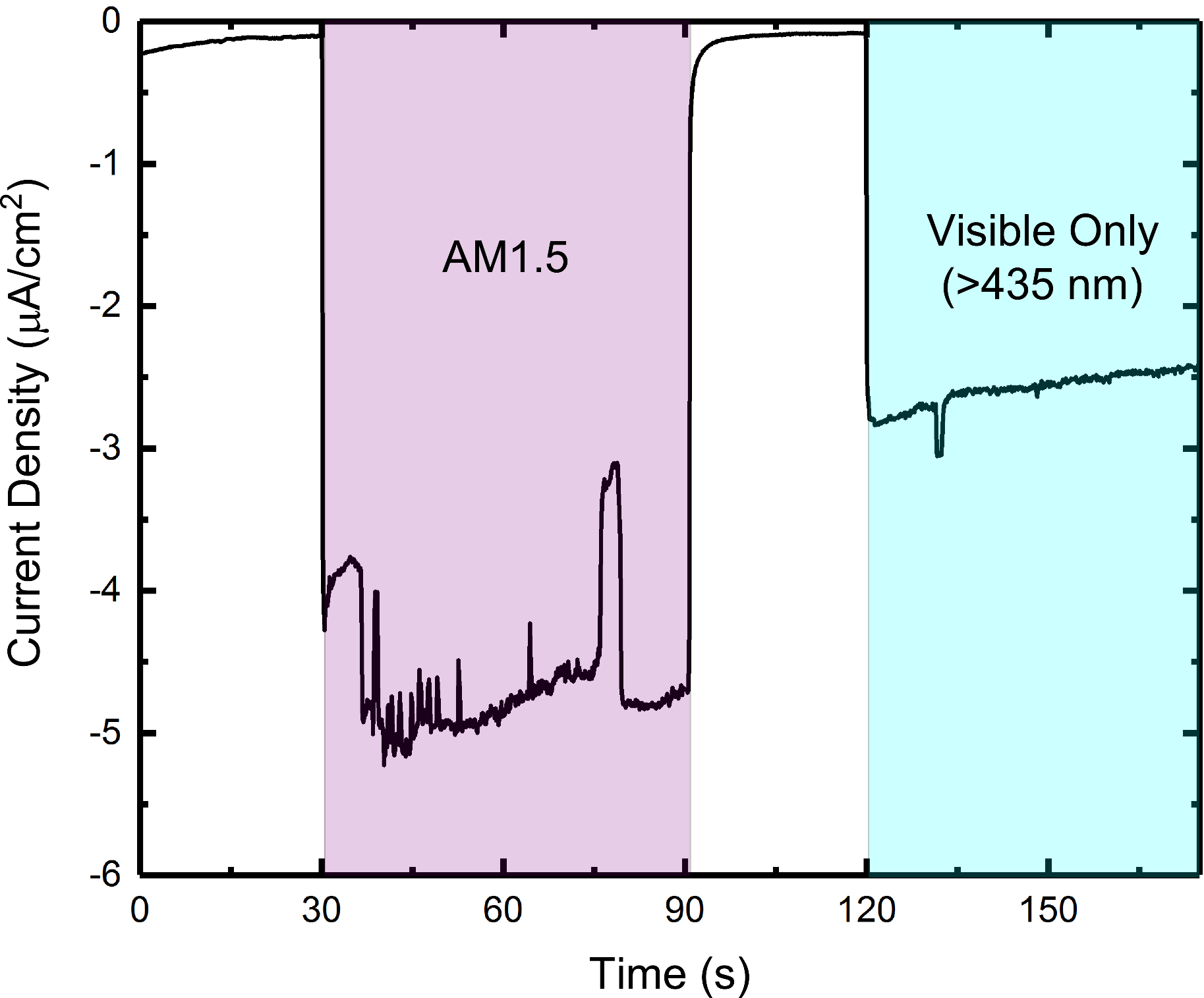
**Figure S2**: Absorbance of 0:1:5, 2:1:10, and 1:2:10 GaxZn1-xOyN1-y powders. Scan was from 850 nm to 350 nm at 1 nm/s.



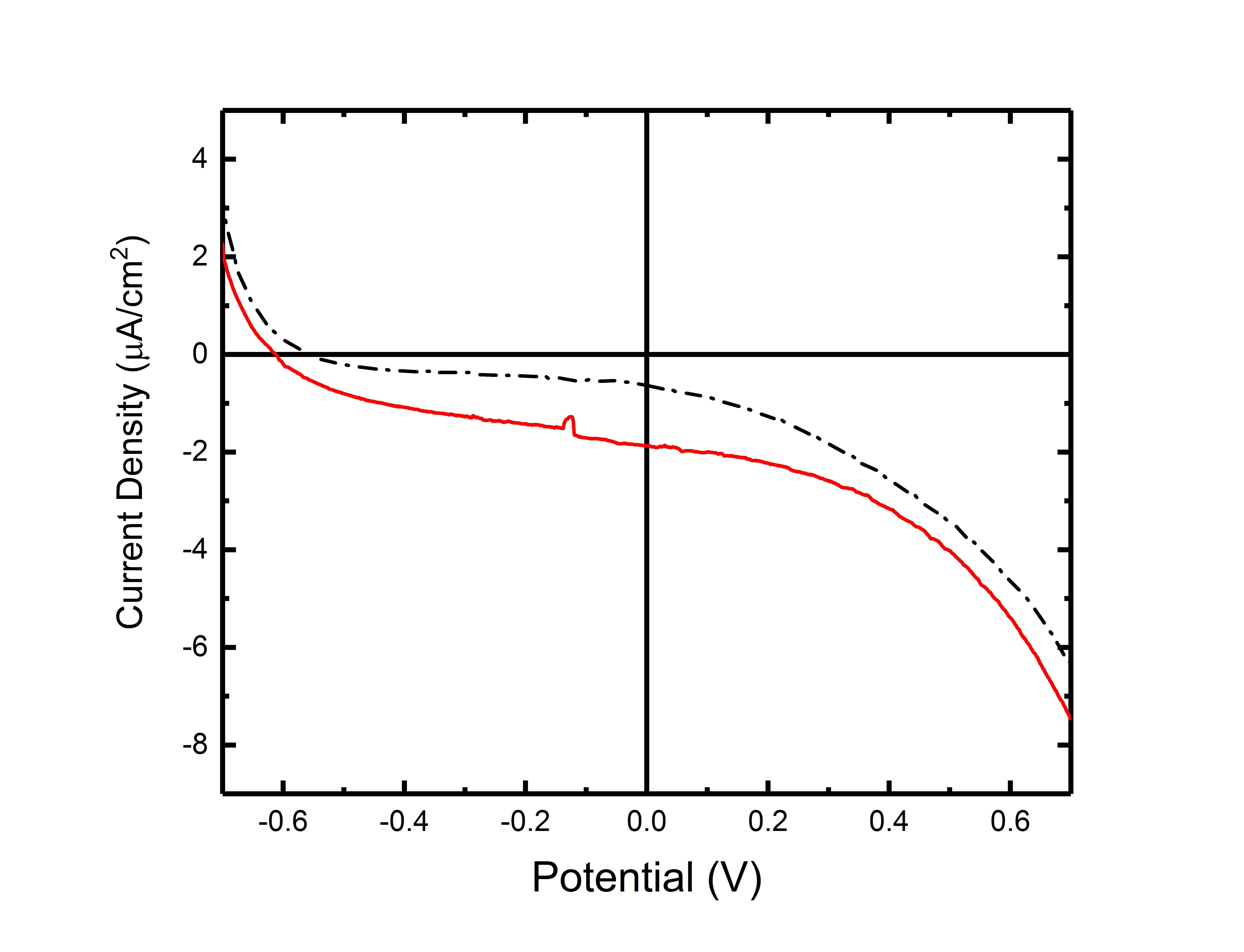
**Figure S3:** X-ray diffraction patterns of 1:1:10 GaxZn1-xOyN1-y powders at different reaction temperatures. Scans were collected at 1o/min with a step size of 0.04o.



**Figure S4**: Current-time measurement of a 1:1:10 GaxZn1-xOyN1-y film (500 oC synthesis temperature) under both visible-only and AM1.5 irradiation. The electrolyte is 0.2M Na2SO3/0.1M Na2SO4, and the potential was held at 0V vs. SCE. Counter electrode was a Pt wire. Dips in the measured photocurrent were caused by fluctuations in the light intensity.

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**Figure S5**: Current-time measurement of a 1:1:10 GaxZn1-xOyN1-y film (500 oC synthesis temperature) under both visible ( > 435 nm) and AM1.5 irradiation. The electrolyte is 0.5M Na2SO4, and the potential is held at 0V vs. SCE throughout. Counter electrode is a Pt wire, and fluctuations of the photocurrent are due to fluctuations in the irradiation source’s intensity.



**Figure S6:** Current-voltage measurement in a 2-electrode system. Counter electrode is a Pt wire, and the electrolyte is 0.5M Na2SO4. Potential was scanned from negative to positive at a rate of 10 mV/s. The red solid line is with AM1.5 irradiation while the black dash-dot line is the dark measurement.



**Figure S7**: A) Hydrogen evolution over time. 0.1g of 1:1:10 GaxZn1-xOyN1-y powder loaded with 1wt% Rh/1.5wt% CrOx in 50 vol% ethanol solution (200 mL total). 100 mW/cm2 AM1.5 irradiation.



**Figure S8**: Oxygen evolution over time. 0.1g of 1:1:10 GaxZn1-xOyN1-y powder in 10 mM AgNO3 (aq) solution with 200 mg of La2O3. 100 mW/cm2 AM1.5 irradiation.

**Table SI.** Comparison of current work and other relevant reported work

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Entry** | **Photocatalyst** | **Light Source** | **H2/O2 Evolution Rate (mol h-1 g-1)** | **Solution** | **Ref** |
| 1 | ZrO2/TaON (IrOx/Cr2O3/RuOx) | 450W High Pressure Mercury | 27.5 / 10 | H2O (pH=3) | 1 |
| 2 | RuO2/  Ga0.88Zn0.12N0.88O0.12 | 450W High Pressure Mercury | 200 / 100 | H2O | 2 |
| 3 | CoOx/BaNb0.5T0.5O2N | 300W Xe ( > 420 nm) | 0 / 300 | 10 mM AgNO3 (aq) | 3 |
| 4 | CrOx/Rh/  Ga0.88Zn0.12N0.88O0.12 | 450W High Pressure Mercury ( > 300 nm) | 12783 / 6626 | H2O | 4 |
| 5 | CrOx/Rh/  Ga0.88Zn0.12N0.88O0.12 | 450W High Pressure Mercury ( > 420 nm) | 800 / 400 | H2O | 4 |
| 6 | CrOx/Rh/  Ga0.45Zn0.55N0.45O0.55 | 300W Xe lamp (AM1.5) | 273a / 30 | 50 vol% EtOH / 10 mM AgNO3 (aq) | This Work |

aAverage rate for the linear increase between hours 1 and 4.

**Table SII**: Comparison of precursor molar ratios and final surface atomic composition of GaxZn1-xOyN1-y samples synthesized at 500 oC as determined by XPS

|  |  |
| --- | --- |
| **Initial Ratio (Ga:Zn:Urea)** | **Final Composition** |
| 0:1:5 | ZnO |
| 0:1:10 | ZnO0.094N0.06 |
| 1:1:5 | Ga0.33Zn0.67O0.75N0.25 |
| 1:1:10 | Ga0.33Zn0.67O0.62N0.38 |
| 1:2:15 | Ga0.22Zn0.78O0.72N0.28 |
| 1:3:20 | Ga0.17Zn0.83O0.79N0.21 |
| 2:1:15 | Ga0.49Zn0.51O0.55N0.45 |
| 3:1:20 | Ga0.58Zn0.42O0.61N0.39 |
| 4:1:25 | Ga0.66Zn0.34O0.55N0.45 |

**References:**

(1) Maeda, K.; Takata, T.; Hara, M.; Saito, N.; Inoue, Y.; Kobayashi, H.; Domen, K. GaN : ZnO Solid Solution as a Photocatalyst for Visible-Light-Driven Overall Water Splitting. *J. Am. Chem. Soc.* **2005**, *127*, 8286–8287.

(2) Maeda, K.; Domen, K. New Non-Oxide Photocatalysts Designed for Overall Water Splitting under Visible Light. *J. Phys. Chem. C* **2007**, *111* (22), 7851–7861.

(3) Kawashima, K.; Hojamberdiev, M.; Yubuta, K.; Domen, K.; Teshima, K. Synthesis and Visible-Light-Induced Sacrificial Photocatalytic Water Oxidation of Quinary Oxynitride BaNb0.5Ta0.5O2N Crystals. *J. Energy Chem.* **2018**, *27* (5), 1415–1421.

(4) Maeda, K.; Teramura, K.; Saito, N.; Inoue, Y.; Domen, K. Improvement of Photocatalytic Activity of (Ga1−xZnx)(N1−xOx) Solid Solution for Overall Water Splitting by Co-Loading Cr and Another Transition Metal. *J. Catal.* **2006**, *243* (2), 303–308.