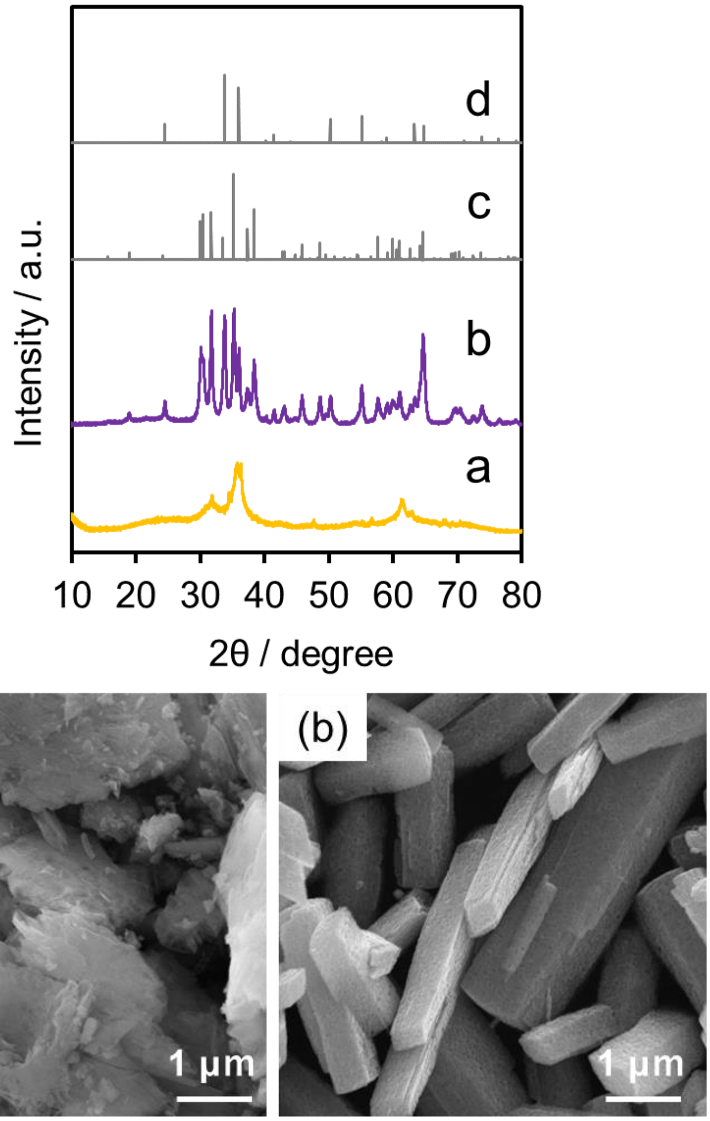


**Figure S1.** (A) XRD patterns and (B) SEM images for (a) δ-$Ga_2O_3$ and (b) K-$Ga_2O_3$. Reference XRD patterns for (c) β-$Ga_2O_3$ (ICDD:00-041-1103) and (d) α-$Ga_2O_3$ (ICDD:01-074-1610) are also presented for comparison in panel (A).

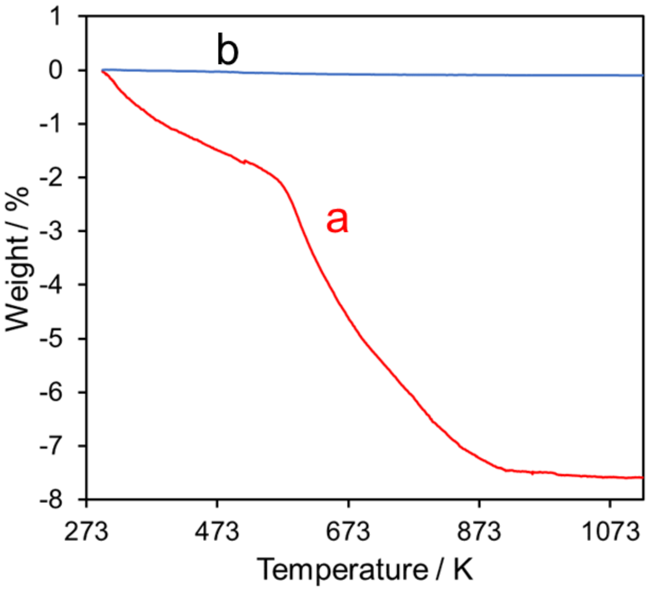


**Figure S2.** TGA profiles for (a) δ-$Ga_2O_3$ and (b) K-$Ga_2O_3$.

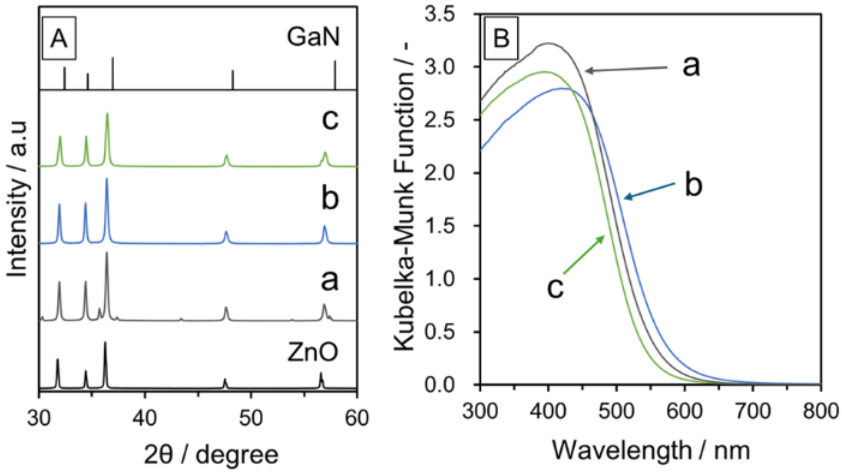


**Figure S3.** XRD and DRS results for GaN:ZnO(δ) samples prepared (a) without addition of Zn, (b) with addition of Zn, and (c) with addition of submicron ZnO particles by heating at 1123 K for 10 h.

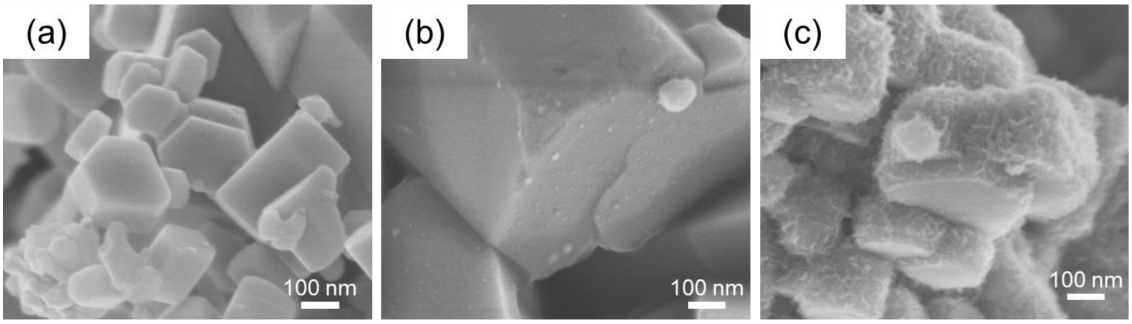


**Figure S4.** SEM images of (a) pristine GaN:ZnO and GaN:ZnO after (b) photodeposition of Pt and (c) additional photodeposition of $MnO_x$.

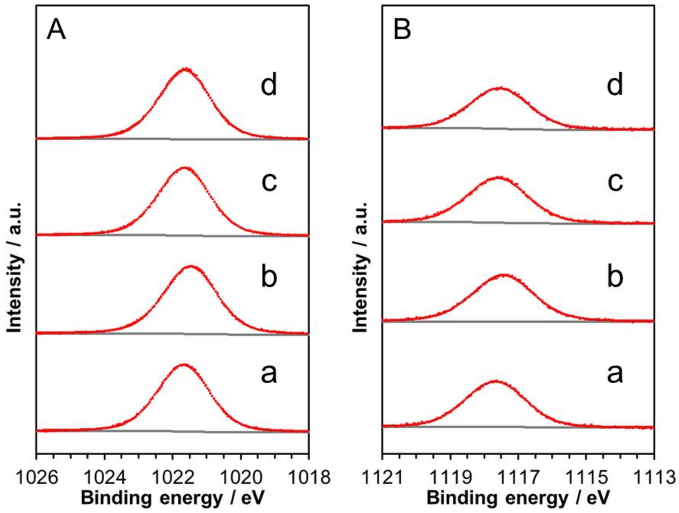


**Figure S5.** (A) Ga2*p* and (B) Zn2*p* XPS spectra for GaN:ZnO synthesized at (a) 1073, (b) 1123, (c) 1173, and (d) 1223 K, respectively.

**Table S1.** The surface atomic ratio of GaN:ZnO(δ) prepared different temperatures estimated by XPS.

| Synthesis Temperature / K | Zn/Ga molar ratio / - |
|---|---|
| 1223 | 2.8 |
| 1173 | 2.0 |
| 1123 | 1.9 |
| 1073 | 1.9 |

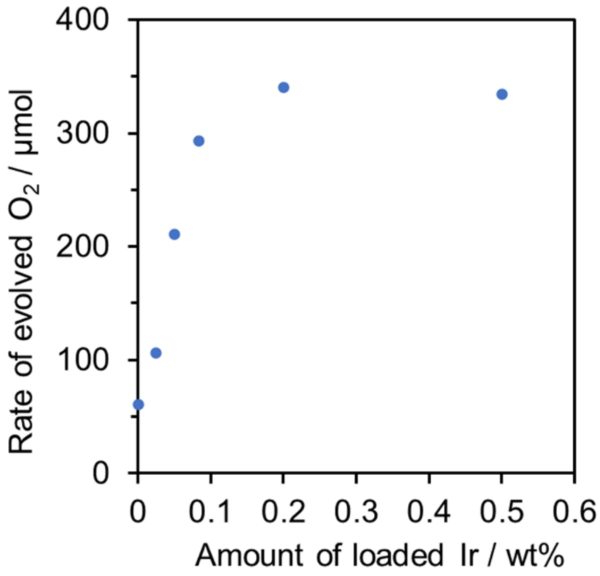


**Figure S6.** Dependence of oxygen evolution activity for GaN:ZnO(δ) synthesized at 1173 K on loading amount of $IrO_x$ (Ir). The reactions were carried out in an aqueous $AgNO_3$ solution (30 mM) under visible light irradiation ($\lambda$ > 420 nm) from a 300-W Xe lamp.

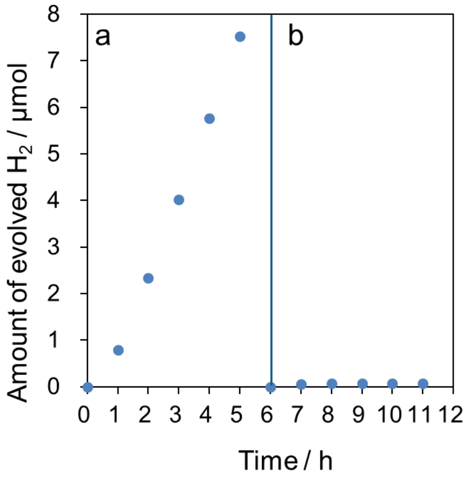


**Figure S7**. Time course of hydrogen evolution from aqueous solution containing (a) 10 mM ascorbic acid and (b) 10 vol% methanol using Rh/Ir$O_x$-loaded GaN:ZnO. Reactions were performed under visible light irradiation ($\lambda > 420$ nm).

**Table S2**. Composition, bandgap energy, and oxygen evolution activity of GaN:ZnO.

| **Ref** | **ZnO / mol%** | **Band gap / eV** | **AQY of oxygen evolution reaction** | **Electron accepter** | **Cocatalyst** |
|---|---|---|---|---|---|
| S1 | 13 | 2.7 | Active[a] | $AgNO_3$ | $RuO_2$ |
| S2 | 17 | 2.7 | Active[a] | $AgNO_3$ | None |
| S3 | 42 | 2.4 | - | - | - |
| S4 | 49 | 2.3 | - | - | - |
| 13 | 49 | 2.3 | 14.3% at 420 nm | $AgNO_3$ | $IrO_2$ |
| S5 | 49 | 2.5 | - | - | - |
| S6 | 51 | 2.2 | - | - | - |
| 17 | 55 | 2.3 | Active[a] | $AgNO_3$ | None |
| 17 | 58 | 2.1 | - | - | - |
| 13 | - | 2.0 | Active[a] | $AgNO_3$ | $IrO_2$ |
| This work | 65 | 2.1 | 11.9% at 420 nm | $AgNO_3$ | $IrO_x$ |
| S7 | 63 | 2.0 | - | - | - |
| S8 | 73 | 2.2 | - | - | - |
| 24 | 79 | 2.2 | - | - | - |
| S9 | 81 | 2.4 | - | - | - |
| S10 | 85 | 2,4 | - | - | - |
| S11 | 90 | 2.8 | - | - | - |
| 11 | 90 | 2.1 | - | - | - |

[a] Capable of evolving $O_2$ but the AQY was not measured.
- indicates no data.

## Supplementary Reference


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