

Supporting Information

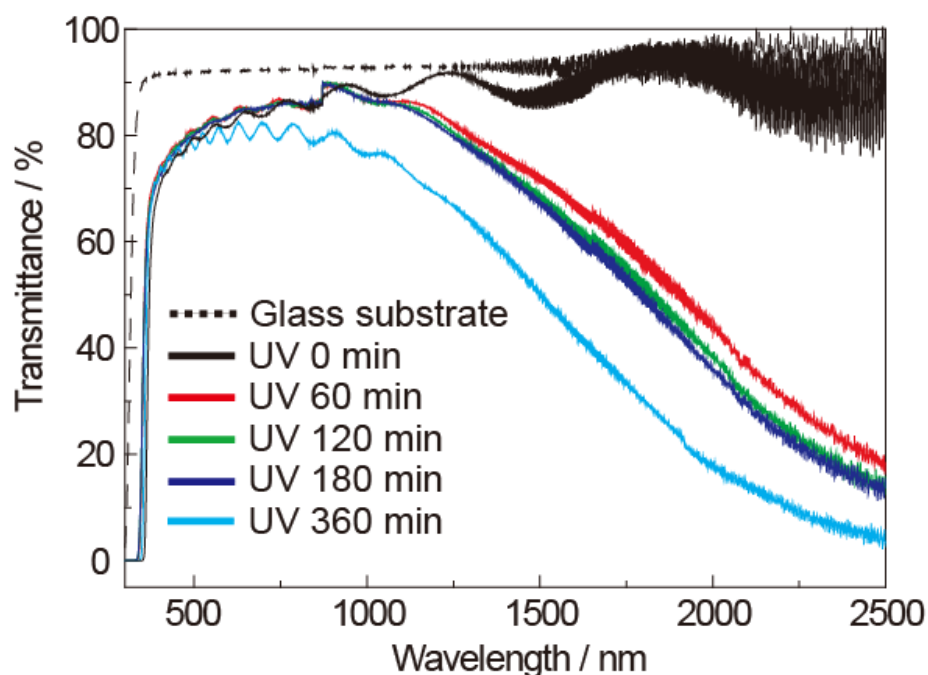


Figure S1. UV-Vis-IR optical transmittance of the UV-illuminated ZnO films.

Figure S1 shows the transmittance spectra of samples treated for various UV illumination times. The average optical transmittance in the visible light range exceeds 70%. The transmittances of the films in this region did not change after UV treatment, whereas that in the near-infrared (NIR) region decreased with the illumination time. The transmittance of the film after 360 min of UV treatment was 5% at a wavelength of 2500 nm. This reduction in the transmittance in the NIR region is attributed to the increasing conductivity (i.e., increasing number of carrier electrons) with UV illumination time; the IR reflection behavior of transparent conductive films originates from the plasma frequency of electrons, as reported in highly metal-ion-doped ZnO films. In the Drude model, the cut-off wavelength at which the IR reflection changes to visible transmission is called the plasma wavelength λ_p of the free-electron material.

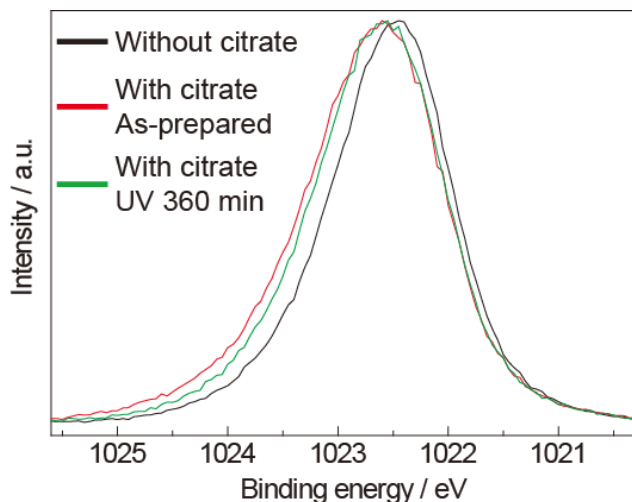


Figure S2. HXPS Zn 2p_{3/2} spectra of ZnO films prepared without citrate, with citrate before UV treatment, and with citrate after UV treatment for 360 min

Figure S2 shows the HXPS spectra of the ZnO films. The Zn 2p_{3/2} peak of the samples prepared with citrate before UV-treatment was slightly wider than the others. Although the peak became narrower after UV treatment, it was still wider than the sample prepared without citrate. The main peak of Zn 2p_{3/2} was attributed to Zn-O bonding, and high energy species were attributed to the other Zinc(II) species, or Zn ion in defective region in the ZnO lattice. These results corresponds those of C1s and O1s, indicating the decomposition of organic impurities by UV-illumination. There were estimated to be COO-Zn bonding because the decrease of sub-peak intensity in C 1s, O 1s and Zn 2p_{3/2} was occurred after UV treatment.

Note that no metal Zn species were observed in the ZnO prepared with citrate due to absence in low-energy species.

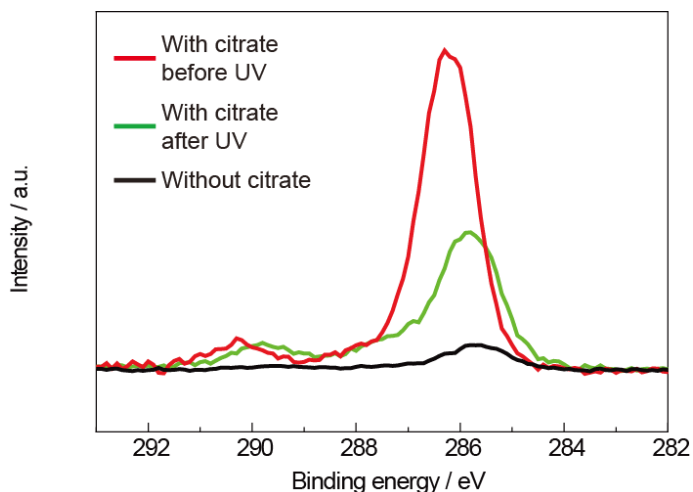


Figure S3. Conventional XPS spectra of C1s core levels for the sample prepared without citrate, with citrate before UV treatment, and with citrate after UV treatment for 360 min.

X-ray photoelectron spectroscopy (XPS) measurements were performed for the ZnO films using a VG scientific sigma probe with nonmonochromated Al-K α (1486.6 eV) radiation and the base pressure of 1.0×10^{-8} Torr.

Table S1 Resistance of the ZnO films after various post-treatment.

Sample	Resistance (k Ω)
As-prepared (dried at 100°C for 1 day)	3.8×10^2
Thermal treatment at 200°C for 4h (without UV treatment)	9.0×10^2
Thermal treatment at 400°C for 4h (without UV treatment)	2.1×10^4
UV treatment for 360 min (without thermal treatment)	0.4
UV treatment + Thermal treatment at 100°C for 4 h	0.69
UV treatment + Thermal treatment at 120°C for 4 h	1.7
UV treatment + Thermal treatment at 140°C for 4 h	15
UV treatment + Thermal treatment at 160°C for 4 h	18
UV treatment + Thermal treatment at 180°C for 4 h	22
UV treatment + Thermal treatment at 200°C for 4 h	13
UV treatment + Thermal treatment at 300°C for 4 h	11
UV treatment + Thermal treatment at 400°C for 4 h	1.8×10^4

The resistance of the films was measured by two-terminal method with a digital multimeter. The distance of the each termination was set at 10 mm. The contact electrodes were not fabricated on the films.

“UV-treatment + Thermal treatment” means that the films were thermal-treated at a given temperature after UV treatment for 360 min.

The difference in the value between two-terminal method and four-terminal method is due to contact resistance on the film and resistance of the equipment in two-terminal method.

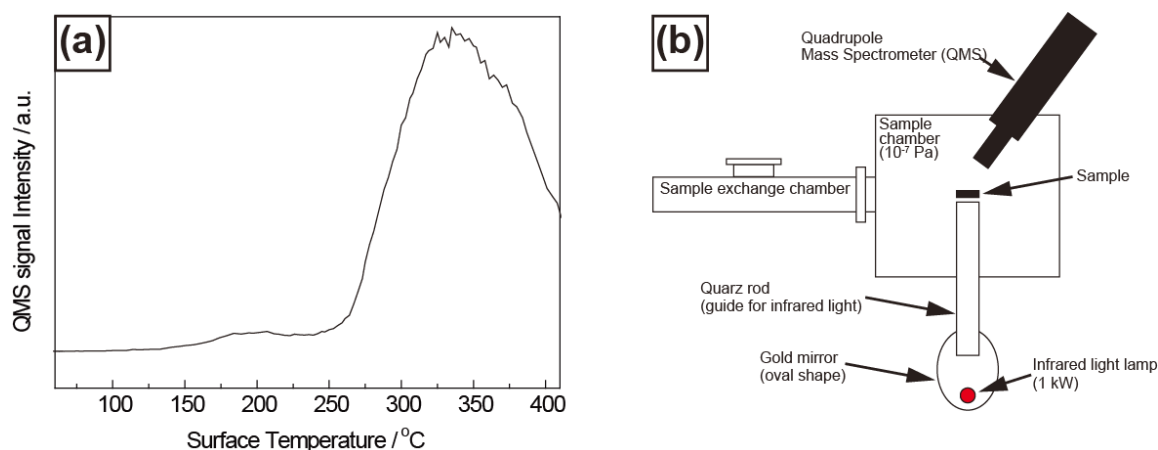


Figure S4. (a) TDS spectra of ZnO films after UV treatment for 360 min. Figure shows a mass fragment (m/z) of 44, which corresponds to CO_2 . (b) The schematic illustration of TDS equipment (EMD-WA1000S/W).

Thermal desorption spectroscopy (TDS) on the samples were performed by EMD-WA1000S/W spectrometer (ESCO, Ltd., Japan). The films on silicon substrate were especially prepared for the measurement. Desorption of CO_2 was observed starting at 225°C in the UV-treated ZnO films. Although the vibrational modes of carboxyl groups diminished in the FT-IR spectra of the films (Figure S1), desorption of CO_2 from the films was clearly observed. Therefore, organic impurities did not decompose completely to water (H_2O) and carbon dioxide (CO_2), but some carbon-related species remained in the films even after UV illumination.