Supplementary Information

Single crystal growth of bulk $InGaZnO_4$ and analysis of its intrinsic transport properties

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The $(InGaO_3)_m(ZnO)_n$ (=IGZO-mn) crystals were grown using the OFZ method under atmospheric pressure. During the synthesis, the vaporization of substances from the molten zone was observed. The vaporized substance deposited on the quartz-tube walls which line the infrared radiation furnace. Figure S1 exhibits the PXRD results of the evaporated and deposited substances. As seen in Fig. S1 these were identified as mainly crystalline ZnO. However, the coincidence between the observed peak at 2θ=33~34° and the diffraction peak of (002) in ZnO is not sufficient, suggesting that a small amount of some oxides consisting of both In and Zn are also present. By qualitative analysis of the PXRD pattern they are presumed to be In-Zn-O-based compounds such as $In_2O_3(ZnO)_{17}$ (see Fig. S1). This belongs to the homologous compounds described as $In_2O_3(ZnO)_n$, the members of which exhibit similar structural properties and hence similar XRD patterns. Therefore we cannot definitively identify individual members and instead have suggested a few such as n=17 as shown in Fig. S1. In addition, $Zn_{1-x}In_xO$ and $ZnO_{1-\delta}$ which also show similar XRD patterns to ZnO could be present. In addition, the Bragg reflection peaks were quite broad (but not amorphous). The presence of multiple phases and broad peaks make it more difficult to perform Rietveld refinement. Therefore, we could not obtain an accurate refinement of XRD data for the evaporated ZnO, but we could at least confirm the evaporation of ZnO and Zn/In containing compounds.

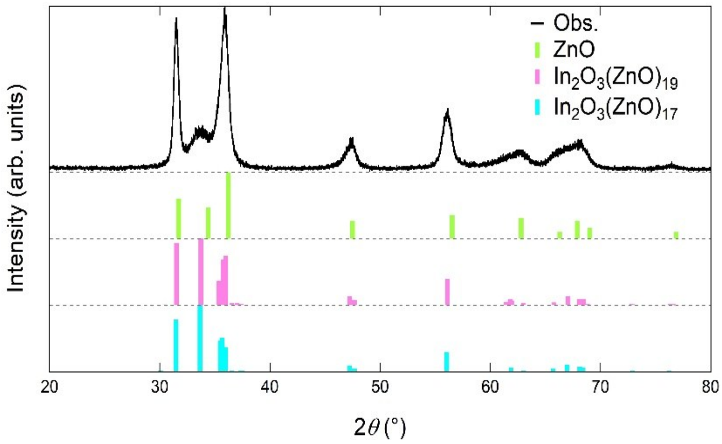

Figure S1 XRD patterns of the vaporized substance deposited on the quartz-tube walls from the molten zone, where Cu-Kα radiation (λ=1.5418Å) was used. The green, pink and blue vertical bars represent ZnO, $In_2O_3(ZnO)_{19}$ and $In_2O_3(ZnO)_{17}$ peaks, respectively.

Figure S2 shows the XRD pattern for ground samples prepared using the conditions for sample #P01Z10. The pattern shows some similarity to (b) in Fig. 2. The relative intensity of the XRD peaks cannot be used for an accurate analysis of the pattern because a number of single crystals remain in the ground sample; e.g., in these crystals, (00l) peaks are relatively stronger than others as seen in Fig. 2, due to their layered structure. However, Fig. S2 at least suggests that the main phase of the as-grown crystals is IGZO-21 (i.e. $In_2Ga_2ZnO_7$). (See Fig. S2).

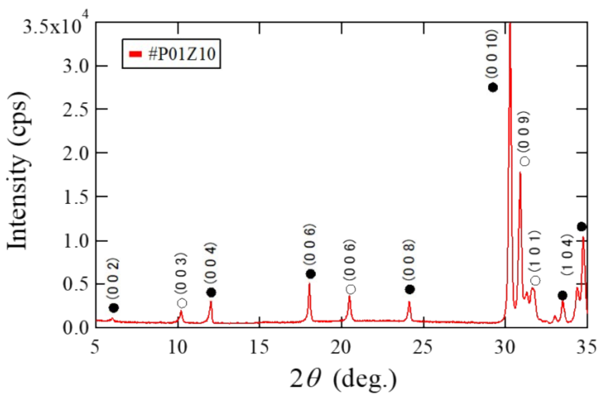

Figure S2 XRD pattern of the grown crystal by the OFZ method under normal pressure, where the growth conditions for #P01Z10 were used (see Table 1 of main article). Black open and solid circles represent the Bragg peaks of IGZO-11 and IGZO-21, respectively.

In order to find the optimal conditions for the single crystal growth of IGZO-11, we investigated the effects of gas-pressure and the ZnO-contents of the feed rods. The results are shown in Fig.S3. For samples prepared using a low-gas pressure, where the ZnO vaporization was not inhibited, IGZO-21 crystals were obtained as the primary product together with Zn-defect rich IGZO-11 crystals (see Fig. S3). On the other hand, samples prepared under pressures of $P>0.8$ MPa combined with a Zn-rich solution, possessed only a small amount of Zn vacancies. In addition, no IGZO-21 impurities were found to be present in the crystal grown by the condition of #P09Z11. In addition, clear Laue spots were observed for crystals grown using the conditions for #P09Z11. This confirms the single crystalline nature of our IGZO-11 samples.

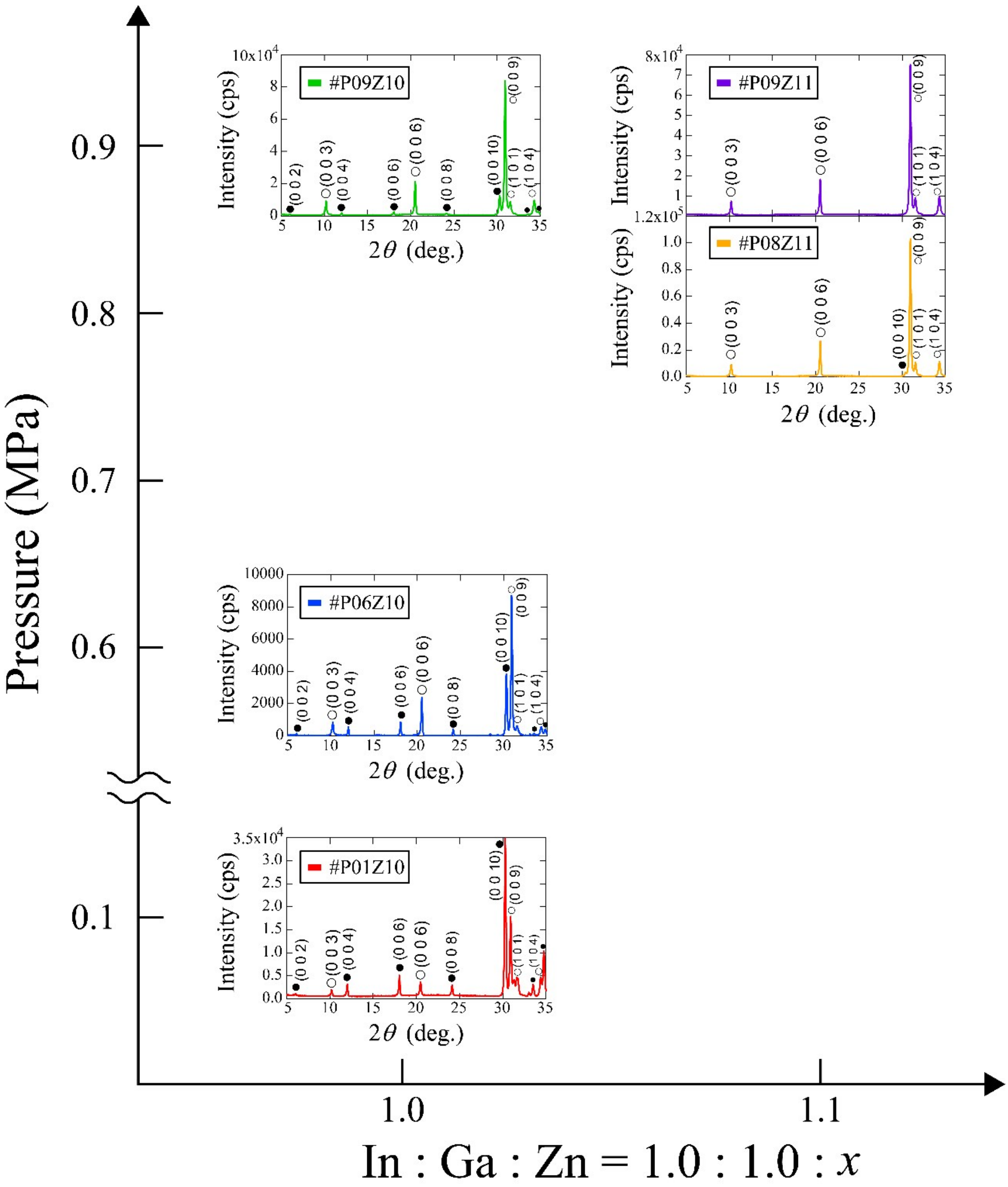

Figure S3 XRD data of crystals grown using various conditions (gas pressure and composition of the feed rod). The details are provided in Table 1. Black open and black solid circles represent the Bragg peaks of IGZO-11 and IGZO-21, respectively. The best conditions to grow IGZO-11 single crystals are those for #P09Z11, where no IGZO-21 peaks are observed. Here, *x* means Zn ratio as In:Ga:Zn=1:1: *x*.