Transparent Thin Film for Energy Harvesting

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Abstract. Amorphous InGaZnO (a-InGaZnO) has already been proved to have good applications in thin-film transistor devices for display application. The a-InGaZnO is n-type semiconductor material and has enormous potential such as transparency, a low temperature fabrication process. Thus, the application of a-InGaZnO to flexible electronics and sensors has been studied. A low thermal conductivity and a low temperature process are merit for a thin film or a flexible thermoelectric module application. In this study, thermoelectric properties of a-IGZO thin film are evaluated. Transparent thermoelectric generator was demonstrated using a-InGaZnO and ITO electrodes.

1. Introduction
Application of thermoelectric conversion of unused waste heat issuing from sources such as the human body and daily lives has recently been studied. It is necessary to investigate new applications such as transparent and flexible thermoelectric devices to incorporate such innovations practically into our daily lives. Therefore, transparent and flexible thermoelectric generators are becoming more attractive for smart windows and wearable systems. To obtain a high thermoelectric property (figure of merit: ZT), a low thermal conductivity is indispensable parameter.

Amorphous InGaZnO (a-InGaZnO) is n-type transparent oxide semiconductor material and has enormous potential such as transparency, a low temperature fabrication process. Thus, the application of a-InGaZnO to flexible electronics for thin film transistor has been studied. The thermal conductivity of the InGaZnO is around 1.0 W/mK due to the amorphous structure [1,2]. Therefore, low thermal conductivity and a low temperature process are merit for a transparent and a flexible thermoelectric module application. Oxide thermoelectric materials can be also used for high temperature applications.

Our group researched thermoelectric properties in the a-InGaZnO thin film [3,4]. We also demonstrated transparent thermoelectric generator (TEG).

2. Experiment
At first, thermoelectric properties of a-InGaZnO film were measured using the sample as shown in Fig. 1. The a-InGaZnO films with thickness of 200 nm were deposited on quartz glass substrate (20 mm × 5 mm, 0.5 mm thickness) using sintered targets (In:Ga:Zn =2:2:1, at.%) by RF magnetron sputtering. We employed an input RF power of 100 W in a mixed Ar and O2 atmosphere at fixed total pressure of 0.6 Pa. After the sputtering process, the a-IGZO samples were annealed. After annealing, AU/Mo electrodes were deposited by electron beam evaporation and formed for ohmic contact. Carrier concentrations were modified by changing the sputtering condition. The Seebeck coefficient and electrical conductivity were obtained in the temperature range from 100 K to 400 K using the
thermal transport option for the physical property measurement system (PPMS, Quantum Design, Inc.). The thermal conductivity was measured using LFA.

![Image](attachment:figure1.png)

**Figure 1.** (a) a-InGaZnO/glass sample to evaluate thermoelectric properties. (b) Thermal transport sample puck (PPMS, Quantum Design, Inc.) with a-InGaZnO sample.

3. **Measurement and results of a-InGaZnO thin film**

Hall mobility was increased with increasing carrier concentration (Fig. 2 (a)). The 13 cm²/Vs was obtained at the carrier concentration of 9.8×10¹⁹ cm⁻³ (oxygen flow ratio was 0% in the sputtering process). Figure 2 (b) shows the values of thermal conductivity. The thermal conductivity was around 1.2 – 1.3 W/mK at 300 K. The thermal conductivity was slightly increased by the carrier concentration due to a heat conduction carrier of electrons. The reported values of poly crystal ZnO or AlZnO film thermal conductivity were around 4 – 6 W/mK [5,6]. Therefore, the amorphous oxide has an advantage for thermoelectric application. Electrical conductivity and Seebeck coefficient were shown in Fig. 3. These results the trade-off relation between electrical conductivity and Seebeck coefficient. ZT value was 0.02 – 0.03 at 300 K and 0.14 – 0.2 at ~800 K (thermal conductivity was assumed 1.3 W/mK). Even amorphous material, the ZT value of a-InGaZnO was almost comparable with InGeO and AlZnO bulk materials owing to the lower thermal conductivity (Fig. 4).

![Image](attachment:figure2.png)

**Figure 2.** Hall motility and thermal conductivity of amorphous InGaZnO sample with different carrier densities.
4. Demonstration of transparent TEG

Transparent thermoelectric device was demonstrated using a-InGaZnO and ITO electrode uni-leg structure (Fig. 5). The sample TEG was fabricated using ITO (155 nm)/Glass substrate with photolithography and etching process. The ITO has a negative Seebeck value (< -10 µV/K). But it was relatively smaller than a-InGaZnO value. Thus we used the ITO as electrode. The 70 pairs of a-InGaZnO and ITO stripes was designed. As shown in Fig. 6, the demonstrated TEG was fully transparent. Output power of the demonstrated device was still small. However, the output power will be improved by optimizing the atomic composition of InGaZnO and the device structure.

Figure 3. (a) Variations of electrical conductivity, Seebeck coefficient and (b) Power factor, as functions of carrier concentration for the different sputtering oxygen ratios of the a-InGaZnO thin films at 300 K (b) PF as functions of carrier concentration at 300 K.

Figure 4. Temperature dependence of ZT values of a-InGaZnO film. Reported values of other bulk oxides were also plotted. [7-10]

Figure 5. Transparent thermoelectric device. (a) Fabrication process. (b) Schematic image of the sample. (c) Optical microscope image of InGaZnO and ITO uni-leg structures.
5. Conclusion
The thermoelectric properties of amorphous InGaZnO thin film were shown. The low thermal conductivity and relatively high Hall mobility was advantages for thermoelectrical applications. Power factor value was still small. But ZT values of amorphous InGaZnO were almost consistent with other poly-crystal oxide materials even amorphous structure. The transparent TEG was investigated using ITO and a-InGaZnO uni-leg structures. For increase the output power, composition of InGaZnO material and TEG structures needs to be further improvement.

References
[1] T. Yoshikawa et al. 2013 APEX 6, 021101
[2] S. W. Cho et al. 2013 J. Nanomat. 909786
[3] Y. Fujimoto, M. Uenuma, Y. Ishikawa, and Y. Uraoka, 2015 AIP advances, 5, 97209,1-6
[4] Y. Fujimoto, M. Uenuma, Y. Ishikawa, and Y. Uraoka, 34th ICT 2015 and 13th ECT 2015
[5] S. Saini, et al., 2015 J. Electron. Mater., Vol. 44, No. 6
[6] S. Saini, et al. 2014 Jpn. J. Appl. Phys. 53, 060306
[7] H. Muta, et al., 2003 J. Alloys Compd., 350, 292–295
[8] M. Ohtaki, et al, 2009 J. Electron. Mater., 38, 1234–1238
[9] K. Fujita, et al.,2001 Jpn. J. Appl. Phys., 40, 4644–4647
[10] D. Berardan, et al, 2008 Solid State Comm., 146, 97

Figure 6. Transparent thermoelectric device of InGaZnO and ITO uni-leg structures. (a) Device image and (b) output characteristics.