論 文 の 内 容 の 要 旨

論文題目　　Biosensors based on amorphous $InGaZnO_4$ films
（アモルファス$InGaZnO_4$薄膜を用いた
バイオセンサに関する研究）

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Amorphous indium gallium zinc oxide (aIGZO) has been vastly industrialized as a key component for transparent thin film transistor (TFT). However, few research has been found yet applying aIGZO to biosensing. The main aim of this dissertation is to study the DNA immobilization method and the possibility to use aIGZO in biosensing. The single strand DNA (ssDNA) immobilization on aIGZO by absorption was examined with a comparison to ITO, which is the first step for many biosensing schemas. The DNA quantification by fluorescence intensity shows that the absorption capacity of aIGZO film to ssDNA is 6.7 times greater than that of ITO. X-ray photoelectron spectroscopy (XPS) and contact angle analysis proved the high DNA adsorption affinity on aIGZO film is related to its high effectiveness to OH attachment (14.7 times higher than ITO).

In this thesis, we also examined the effect of hydroxyl radical and thermal crystallization on DNA immobilization to aIGZO thin film. Evidenced by XPS and fluorescence image, hydroxyl radical concentration in the solution is positive correlated to DNA immobilization. On the contrary, by comparing with crystalline IGZO, thermal annealing has a negative effect on the immobilization. The OH affinity on aIGZO is 56 times higher than crystallized film, and hence, its DNA absorption quantity is 24 times higher than crystallized one. Amorphous thin film, consequentially, surpasses the thermal crystallized film on DNA immobilization, and is more suitable for biosensor. This thesis presents three practical methods for controlling DNA immobilization on aIGZO thin film surface by changing OH solution concentration and thermal annealing and the oxygen partial pressure at film growth. Reusability of the surface was also evaluated through physical restoration. A physical reusable schema by introducing sonication with proper pH (6.8) washing buffer was developed. Through optimization, 67% of the film surface can be reused for DNA biosensing.

Besides the DNA immobilization method, the most significant findings in this

research relate to new property of aIGZO thin film that the OH affinity on its surface overpasses other materials and is controllable. This property is also the primary reason for using aIGZO in biosensing, because the OH on the thin film surface determines the DNA quantity, which can be absorbed. To make use of the outstanding OH affinity on aIGZO thin film, and to facilitate the evaluation of the surface OH quantity for biosensing, fluoresce, impedance, resistance, contact angle and DNA based OH sensors target at strong alkaline range (above 100 mM) were invented. The resistance OH sensor invented which has the maxima resolution sensitive to 1.5 mM at the target strong alkaline region can be categorized as a fine evaluation method. Proper method can be selected accordingly. Other methods such as XPS or secondary ion mass spectrometry (SIMS) and mass spectrometry (MS) methods are the scientific methods to evaluate OH quantity with high sensitivity. But all of them need high specific equipment, which is too large with no portability. There is no portable method to evaluate OH concentration directly except our OH sensors. Most time pH sensor is used instead. But the pH sensor is targeting at the counter part of OH, and in strong alkaline range it shows no sensitivity. Therefore, the OH sensors invented based on aIGZO are the promising solution for solving practical problem to evaluate strong alkaline solution, meanwhile enrich the usage of the aIGZO in sensing.

Finally, biosensing applications based on aIGZO thin films are proposed in this work. Impedance DNA sensor based on aIGZO thin film enables optical detection and electrical detection on the same platform. It is possible to enable a multi-detection biosensor as a self-verifiable DNA sensor. We successfully realized the electrical sensing on the multi-detection platform by impedance to DNA length. According to our experiment the limitation of the impedance and fluorescence DNA sensor are both around 40 μM. This may because of the limitation from the measurement apparatus. However, the sensitive resolutions are different: 7.7 μM from the fluorescence sensor, 3.5 μM from the impedance sensor at 40~67 μM range. To apply the multi-detection biosensor based on impedance and fluorescence for DNA sequencing, the impedance signal could be used to sense the length of the DNA, and the fluorescence signal could be used to determine the type of the nucleotide. TFT biosensor was also proposed with fabrication and simulation practice. The simulation result demonstrates the possibility to implement fluorescence DNA biosensing combining with the current sensing signal. It may enable the optical and electrical multi-sensing platform by introducing transparent TFT based on aIGZO. All applications mentioned above make use of the aIGZO thin film surface characteristics and extends its possibility for usage in biosensing.