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主 論 文 の 要 旨

論文題目　**Designed Synthesis of ZnO Nanowires in Hydrothermal Process and Their Photoelectronic Nanodevice Application Towards Biomolecule Analysis**
(ZnO ナノワイヤ水熱合成の精密設計と生体物質の光・電気解析ナノデバイスへの展開)

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論 文 内 容 の 要 旨

Recently, zinc oxide (ZnO) nanowires have drawn a great deal of interest due to their outstanding physical properties, various approaches of achieving, feasibility of further modified or heterojunctions with other materials, controllable morphologies, and biocompatibility. Possessing of these significant properties, ZnO nanowire is a potential candidate as core component embedded in biosensor. Improvement in genomic and molecular methods make it possible to identify both genetic and epigenetic mutation through microinvasive or noninvasive achieved liquid biopsies (such as blood, urine et. al.). A high-throughput, high-sensitivity and individualized detection of biomarkers in the liquid biopsy is urgently required. Technological advances in nanomaterial and nanofabrication promote the miniaturization and integration of ZnO individual nanodevices enabling realization of the requirement. However, the realization of industrialized applications of ZnO nanowire-based devices is still hindered by several reasons. First of all, the gap between the require of simply cost effectiveness on synthesis of large aspect ratio ZnO nanowire and the presented solutions, leaving most of the architectures remain stuck at nanowires/nanorods array-based, which sets a limitation of taking full advantage of ZnO nanowire in the 1-D nanostructure. Secondly, a fair amount of researchers flinch at the complexities in constructure of individual nanowire device. Furthermore, it is difficult for a mere ZnO nanowire to keep stability in various complicated environments, particularly in aqueous surrounding. It is concluded that serious obstacles need to be overcome to realize wide applications of ZnO nanowires.

This thesis has been devoted to study on designed synthesis of ZnO nanostructures towards photoelectronic nanodevice application for biomolecule analysis in liquid biopsy. The literature research in Chapter I mentions that electrical, optical and mechanical properties of ZnO nanowires are strongly depending on the structures like morphology, crystallinity and surface characteristics, designed synthesis of nanowires are in high demand to further develop the research of ZnO nanowires. In addition, exploring the electron transport property and stability reinforcement of single ZnO nanowires enables novel design of minimized electrical nanodevice. From chapter II to chapter IV, I exhibit the content of my thesis starting from designed synthesis of ZnO nanostructures towards photoelectronic nanodevice application for biomolecule analysis in liquid biopsy.

Chapter II presents a seed layer engineering strategy to control the morphology and intrinsic crystallinity of ZnO nanowires via a hydrothermal method. The process in which nanowires grow from prepared seed template layer, ammonia is added in growth system to promotes growth of nanowires. Although the effect of ammonia on the nanowire growth has been intensively investigated, an influence for the seed layer, which governs the initial nanowire growth, is rarely discussed. Here current part of this thesis demonstrates that ammonia strongly affects the seed layer as well as the following nanowire growth. When increasing the ammonia concentration, the nanowire density first increases and then decreases while the nanowire growth rate keeps increasing. Experimental results and thermodynamic calculations as to the initial growth process reveal that the transformation of seed layer induced by ammonia prior to the nucleation critically determines the nanowire density and thus also influences the following nanowire growth. Present results highlight the critical importance to discuss the variation of seed layer in ammonia-contained hydrothermal synthesis and suggest a novel seed engineering approach for tailoring the ZnO nanowire growth, and even physical property.

In chapter Ⅲ , I describe a manageable procedure to embed individual ZnO nanowire in device, and explore the electron transport property of it. Based on the results in Chapter II, sparse ZnO nanowire array with aspect ratio of ca. 120 and growth rate of 1 □m/h is synthesized by controlling density of seeds by ammonia and polyethyleneimine at initial stage of nanowire growth, and prolong the growth period by refreshing nutrient solution. The spatially-separated nanowires were cut off from growth substrate unbrokenly, and thus facilitate to construct a single-nanowire device by photolithography. The device exhibited a linear current-voltage characteristic associated with ohmic contact between ZnO nanowire and electrodes. The device further demonstrated reliable photoresponse with an I_{UV}/I_{dark} of ~100 to ultraviolet light irradiation.

In chapter Ⅳ, I propose the establishment of ZnO-based individual nanodevices with high stability and sensitivity towards biomolecules analysis. The implementation approach is that, atomic layer deposition (ALD) technology boosted constructure of ZnO/ Al_2O_3, ZnO/ SnO_2, and ZnO/ NiO core-shell nanowires. Nanodevices embedding the core-shell nanowires were responded to miRNA 21 in aqueous environment. Thermal annealing was implemented to improve nanowires stability in aqueous environment. Formation of core-shell nanostructures, on the one hand, enhanced the electrical response of ZnO nanowire to biomolecular potentially. On the other hand, thermal annealing caused diffusion of deposited metal oxide layer on ZnO nanowire surface increased stability of nanowire. Comparison of response among nanodevices with different typed of deposited metal oxide layer assisted to reveal the response mechanism. A mature research achievement on core-shell nanodevices is expected to realize selective fingerprinting of miRNAs by material-dependent nanosensors arrays.

Finally, I summary the results and highlights of this thesis. Based on the current result, it is predictable that combination between high density circuit integration of metal oxide nanowire devices for data acquisition and machine learning for data analysis in the fields of physical sensing, chemical detection, and medical diagnosis, would play a significant role in the process of build Internet of Things (IoT) society.