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主 論 文 の 要 旨

論文題目 Nanowire Structures Embedded in Microfluidic Channels for Biomolecules Manipulation and Analysis （ナノワイヤ構造体を組み込んだマイクロ流体チャネルによる生体分子の操作と分析）

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論 文 内 容 の 要 旨

This thesis proposes the biomolecules manipulation and analysis based on nanostructure and nanomaterial embedded in the microfluidic channels. The strategies to achieve our goal can be provides in three parts. Part I is the nanopore DNA sequencing, which describe in chapter 2 and 3. Part II describes the nanowire growth techniques in chapter 4-6. In part III, the applications of nanowire structure embedded in microfluidic channel for the biomolecules manipulation and analysis are described in chapter 7-10.

In Chapter 2, the Electrode-embedded Nanopore (EN), which is the next generation DNA sequencing, has been introduced. We fabricated 0.8 nm Au electrode gap by self-breaking electromigration technique on Si substrate. The tunnelling current was detected during DNA oligomers translocate through the nanopore device.

In chapter 3, micropillars were putted in front of nanopore device to elongate λ DNA (48.5 kbp) before the DNA translocation at the 50 nm nanopore on Si substrate. The transverse electric field was applied at the electrode to control the translocation velocity of the DNA molecule. The DNA translocation was detected by the blocking current measurement during DNA pass through the nanopore device. However, it is still difficult to elongate long DNA molecule because the different level of blocking current from λ-DNA folding were detected. Thus the separation and elongation of the DNA molecules is required.

According to the theoretical and simulation study, the pillar size should have similar with the Kuhn length of DNA molecule (~100 nm), consequently bottom up nanowires were proposed to elongate long DNA molecule. Then, the bottom up nanowires by vapor liquid solid was introduced in chapter 4. The controlling of ITO nanowire resistivity was proposed by varying the Sn component in In2O3 target material and incorporated at Au catalyst via VLS mechanism. The resistivity of ITO nanowire which growth through Au catalyst differ from ITO thin film due to the nucleation probability of liquid-solid (LS) interface. We found that the resistivity from our approach (~10-4 Ω.cm) is lower than the ITO nanowire

from the previous works.

In chapter 5, I investigated and optimized the Vapor Liquid Solid (VLS) nanowire growth techniques to control the crystal phase of metal oxide material. Under the precisely control of Sn/In vapor flux, the crystal phase of nanowire could vary from Rutile phase (SnO2) to Fluorite phase (ISO) and Bixbyite phase (ITO) due to the nucleation at LS interface via VLS technique.

In chapter 6, the anisotropic of ZnO nanowire growth by hydrothermal method was introduced. We found that the critical concentration of Zn ionic species play an important to control the growth direction of nanowire. If the concentration of Zn ionic specie higher than the critical concentration, the sidewall growth in a-plane (10-10) will be observed consequently, the aspect ratio of nanowire decreased.

In chapter 7, we have demonstrated self-assembly nanowire structure, which is bottom-up approach nanostructure, embedded in microchannel as spot-array structures for single molecule DNA manipulation. The T4 DNA (166 kbp) could elongate by nanowire spot array structure more than 80 % of its contour length. We also demonstrated the DNA separation by nanowire spot array and random growth nanowire in microfluidic channel. Consistent with our results, spot-array nanowire structures have a feasibility to integrate with other nanostructures to detect biomolecules such as nanochannel or Electrode-embedded nanopore.

In chapter 8, three-dimensional (3D) nanowires structure devices have been developed for DNA electrophoresis to overcome the limitation of separation range and save the analytical time of DNA molecules. The 3D nanowire structures can be synthesized under vapor-liquid-solid (VLS) technique by decorating Au catalyst on the nanowires and growing the branch of nanowires. This method allows researchers to control the pore size between nanowires by increasing the number of nanowire growth and to determine a suitable growth time for the analytical range of DNA molecules. From our results, the rigid network structure allows researcher to analyze the DNA molecules from 100 bp & # 8211; 166 kbp within 13 s under the applied DC electric field, which are much wider and faster than any existing methods.

In chapter 9, the highly dense 3D nanowire structures have been used for small biomolecule separation such as DNA, protein and RNA. We could separate 6 fragments of small DNA molecule (50 - 1000 bp) within 50 s, and the 5 mixture of SDS modified - proteins (20 - 340 kDa) could separate within 5 s. Interestingly, 0.1 - 1 kb RNA ladder was separated within 25 s by the highly dense 3D nanowire structures devices. We could separate the small biomolecules because the controlling of 3D nanowire pore size by varying the nanowire growth as a cycle. Based on our concept, we could enhance the analytical range and save the analytical time by using 3D nanowire structure as a sieving material embedded in microfluidic channels.

In chapter 10, self-assembled nanowires was growth in microfludic channel as filter for DNA filtration. We found that λ-DNA could filter from T4-DNA within 1 s by using self-assembled 3D nanowires structure. We confirmed the filtration mechanism by observing

the single DNA molecule migration behavior of λ-DNA and T4-DNA. We found that λ DNA could pass through the nanofilter while T4 was could not enter to the nanowire area. Consequently, 3D nanowire could be applied as a filter for biomolecule filtration in short time with a high throughput.

This thesis provides the new approaches, which has a potential to reduce the analytical time and cost for biomolecule analysis, by using bottom up approach nanowire structure as sieving matrix material embedded in microfluidic channel. Based on the above results, we believe that nanowire structures have a potential to apply and integrate with nanobiodevices and microfluidic system for analytical chemistry, biomedical and clinical applications.