EFFECTS AND APPLICATIONS OF INTERACTION BETWEEN SILICON NANOWIRE/CARBON NANOTUBE AND BIOLOGICAL MOLECULES

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ABSTRACT

Nanomaterials (NMs) have infiltrated the field of biology and medicine with applications including biosensing, bioimaging, tissue engineering, and drug delivery. On the other hand, biomolecules, such as nucleic acids, proteins and even living virus, are being used to control both the synthesis and assembly of NMs. Therefore, it is fundamentally important to understand the interactions between NMs and biological systems from molecular to cellular levels in order to control and manipulate these interactions for biological applications and for development of biomolecule-NM composites with novel functions. These researches are the pillars of the emerging field of nanobiotechnology. This study herein mainly focuses on two major topics of nanobiotechnology: investigation on protein-NM interactions (including Chapter Two and Chapter Three), and biosensing applications of NMs (including Chapter Four and Chapter Five). In addition, preliminary results on the synthesis and characterization of protein conjugated nanocomposites are also presented.

The first part of this study presents the investigation of the effects of NMs with and without surface functionalizations, including SiNWs and CNTs, on biochemical reactions catalyzed by three important enzymes in biology, namely, restriction endonuclease, DNA polymerase and RNase A. The results have shown that interactions between NMs and enzymes can inhibit enzyme activity, probably due to the adsorption and/or chemical reactions between the functional groups on NMs and enzymes. The inhibition effect on enzyme activities was increased in the order of

SiNW-H > SiNW-COOH > SiNW-SiO_2, CNT-COOH > pristine CNT, CNT-COOH > SiNW-COOH and SWCNT > MWCNT. Both the curvature of NMs and surface functional groups played important roles in influencing the function, structure and activity of RNase A. A larger surface area of contact and a higher surface potential will strengthen the protein-NM interaction. Our study clearly shows that the enzyme is less active on CNT surface than in free solution and the activity decrease further on larger nanotubes. Due to less carboxylic groups on SiNW surface than on SWCNT surface, SiNWs showed a weaker protection effect versus SWCNTs, even though SiNWs have larger diameters than SWCNTs. MWCNTs and SWCNTs showed different tendency in influencing DNA polymerase and RNase A, which is consistent with previous reports that the structure, function, activity and stability of proteins can be strongly influenced by both the surface chemistry of the NM and its curvature upon their interactions, but in a protein-dependent manner.

Though nano-devices basing on SiNW field effect have been intensively investigated, biosensing applications of SiNWs are still limited and at their early stage. It has been shown previously that Ag nanoparticles on roughened surfaces can stimulate strong surface-enhanced Raman scattering (SERS) signal upon interaction with targeted molecules. However, the preparation of metal substrates is the key issue that dictates signal intensity and reproducibility of SERS. We herein fabricated vertically aligned SiNW arrays using metal nanoparticle induced chemical etching method, and thereafter decorated Ag nanoparticles to SiNWs using galvanic displacement reactions.

This vertically aligned SiNW array with Ag nanoparticles decoration is proven to be an ideal substrate which can generate large and predictable enhancements for SERS, since the Ag nanoparticles are pre-organized as aggregates in a uniform manner on the SiNWs. Two major effects are involved in the enhancement of Raman signals: electromagnetic effect associated with dipolar resonance occurring on the metal surface, and chemical effect from scattering process induced by chemical interaction between molecules and metal surfaces. DNA nucleosides, single-strand DNA, double-strand DNA and proteins could give specific and good-quality SERS spectra. It is thus expected that label free microarrays using SERS detection can be fabricated using patterned SiNW substrates. To this perspective, an effective procedure for the fabrication of vertically aligned patterned SiNW arrays with micro-sized features is thus developed by combining single-step photolithography and metal nanoparticle induced chemical etching at room temperature. The patterned SiNW arrays can be used as a platform for specific biodetection based on SERS, where the inherent "fingerprint" SERS spectra allow for the differentiation of closely related biospecies.

At last, preliminary results on the covalent conjugation of mouse IgG to SiNW and CNT surface through carbodiimide reactions between carboxyl on NMs and amine in protein have also been presented. XPS and ATR-FTIR measurements were carried out to validate the presence of a covalent linkage between NMs and proteins. The visualization of proteins on the surface of NMs was also successfully achieved using atomic force microscopy (AFM).

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LIST OF ABBREVIATIONS

1D	one dimensions
3D	three dimensions
AFM	atomic force microscopy
ATR-FTIR	attenuated total reflection Fourier transform infrared
BSA	bovine serum albumin
CCD	charge-coupled device
CD	circular dichroism
CMOS	complementary metal oxide semiconductor
CNT	carbon nanotube
CNT-COOH	carboxylic groups functionalized carbon nanotube
CPMV	cowpea mosaic virus
CV	cyclic voltammetric
CVD	chemical-vapor deposition
dATP	deoxyadenosine triphosphate
dCTP	deoxycytidine triphosphate
dGTP	deoxyguanosine triphosphate
DNA	deoxyribonucleic acid
dsDNA	double-strand DNA
dTTP	deoxythymidine triphosphate
DWCNT	double-walled carbon nanotube
EDC	1-ethyl-3-(3-dimethyl-aminopropyl) carbodiimide
EDS	energy dispersive X-ray analysis
EIS	electrochemical impedance spectroscopy
FET	field effect transistor
fM	femto-molar
FRET	Forster resonance energy transfer
HaCaT	human keratinocytes
HEK	human epidermal keratinocytes
HepG2	hepatocellular carcinoma cell
HF	hydrofluoric acid
HSP60	heat shock proteins
HTA	hydroxylterephthalate
IgG	immunoglobulin
Ikk	IκB kinase
MS	mass spectrometry
MEPs	mean free paths
MWCNT	multi-walled carbon nanotube
NHS	N-hydroxysuccinimide
NM	nanomaterial
NT	nanotube
NW	nanowire

NP	nanoparticle
OAG	oxide-assisted growth
OEG	oligo(ethylene glycol)
PAA	poly(acrylic acid)
PC12	pheochromocytoma cell
PCR	polymerase chain reaction
PNA	peptide nucleic acid
pRNA	DNA-packaging RNA
PSA	prostate specific antigen
QD	quantum dot
RNA	ribonucleic acid
RNase A	ribonuclease A
ROS	reactive oxidizing species
RT	room temperature
SEM	scanning electron microscopy
SERS	surface-enhanced Raman scattering
SiNW	silicon nanowire
SiNW-H	hydrogen-terminated silicon nanowire
SiNW-SiO_2	as-grown silicon nanowire
SiNW-COOH	carboxylic groups functionalized silicon nanowire
ssDNA	single-strand DNA
STV	streptavidin
SWCNT	single-walled carbon nanotube
TA	terephthalic acid
TEM	transmissions electron microscopy
TMV	tobacco mosaic virus
TS	tectosquares
UV	ultraviolet
VFET	variable field effect transistor
VLS	vapor-liquid-solid
XPS	X-ray photoelectron spectroscopy