ADVANCES IN NANOTECHNOLOGY

Volume 17

Zacharie Bartul
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Editors

NOVA

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nova
science publishers
www.novapublishers.com

ISBN 978-1-53611-004-3

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nova
science publishers
New York

Library of Congress Cataloging-in-Publication Data

ISBN: 978-1-53611-004-3
ISSN: 2159-1490

Published by Nova Science Publishers, Inc. † New York

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PREFACE

Nanotechnology is the study of the controlling of matter on an atomic and molecular scale and is also very diverse, ranging from extensions of conventional device physics to completely new approaches based upon molecular self-assembly. This book gathers and presents data on nanotechnology. Chapter One examines the role of atomic particles and nanoparticles in catalysis. Chapter Two reviews the preparations of nanoporous microelectrodes, their physical and electrochemical characterization, their electrochemical behaviors, and their promising applications. Chapter Three studies controllable growth methods for device applications of gold nanorods. Chapter Four provides an overview of various antibody-nanoparticle conjugating modalities and their potential clinical application. Chapter Five reviews the numerous methods available to fabricate delivery systems depending upon the physicochemical properties of active ingredients and the polymers. Chapter Six discusses recent achievements in the field of development of different photoconverters (photodetectors and solar cells) based on nanoheterostructures with quantum dots of germanium on silicon.

Chapter 1 - The role of atomic particles and nanoparticles in catalysis continues to attract extensive investigations from both fundamental and industrial perspectives. Recently, the authors added the novel atomic negative ions to the study of catalysis at the atomic and nano scales. The fundamental mechanism of negative-ion catalysis is the weakening or breaking up of moleular bonds in the transition state by the negative ion. Here the complex angular momentum method is used to generate atomic negative ions from selected complex atoms through the dramatically sharp resonances, manifesting electron attachment, that characterize the calculated low-energy

electron elastic total cross sections (TCSs). The atomic Au TCSs are used as the benchmark for selecting the appropriate atoms. Ramsauer–Townsend (R-T) minima, shape resonances (SRs) and binding energies (BEs) of the resultant anions formed during the electron-atom collisions are extracted. The striking features of these TCSs, exploited in this chapter, are the appearance of a dramatically sharp resonance with large BE in the ground state second R-T minimum and a broad deep R-T minimum in the excited state TCS; thus providing an excellent environment and mechanism for breaking up molecular bonds in new molecules creation from atoms as well as in anionic nanocatalysis. Selected generated negative ions are demonstrated in various applications, including in the dynamic negative-ion nanocatalysis of various chemical reactions: 1) Catalysis of water to peroxide, 2) Negative-ion catalysis of light, intermediate, and heavy water to corresponding peroxides, 3) Methane oxidation to methanol without CO_2 emission, 4) Multiple atom super nanocatalysts, 5) Single atom multifunctional nanocatalysts and sensors, 6) Tunable catalysis of water to peroxide with atomic catalysts and 7) Novel mechanism for creating long-lived (metastable) atomic negative ions. The authors conclude by discussing prospects of using some of the gaseous multiply functionalized anionic nanocatalysts to resolve the problem of deep oxidation in the partial oxidation of methane to methanol.

Chapter 2 - Nanoporous microelectrodes are a new class of very promising techniques for a variety of applications. They combine the advantages of regular microelectrodes and nanoelectrode arrays because unique open nanostructures impart unusually high electrochemical areas and electrochemical activity while keeping the merits of traditional microelectrodes. This chapter reviews the preparations of nanoporous microelectrodes, their physical and electrochemical characterization, their electrochemical behaviors, and their promising applications. Of numerous fabrication methods, the mechanical abrasion method is used to mechanically attach powdery nanoporous materials to electrode substrates. The liquid crystal template method enables the deposition of metals within a template onto the substrates, resulting in the formation of ordered nanoporous metal layers. Electrochemical deposition and dissolution of an active metal onto an inert electrode substrate are a green-chemistry process which creates very thin surface nanoporous layers on the substrate. All of the three methods are convenient and versatile without requiring expensive equipment and complicated processes. Based on the polarization theories of porous electrodes, the utilization of nanopore surfaces is strongly dependent upon the ionic strength of reaction media, pore geometric parameters such as pore

diameter and length, and the kinetics of electrode reactions. High electrode utilization has been demonstrated for several sluggish electrode reactions in high ionic strength media. The nanoporous structures provide unique environments for reactant species to increase their interactions with electrode surfaces and to prevent reactive intermediates from escaping back to bulk solutions, resulting in the increases of faradaic signals. The nanoporous microelectrodes have demonstrated their prospects for a wide range of applications, especially for energy storage conversion, electrochemical analysis and sensors, and neural stimulation and recording. Further studies and development of this class of electrodes will boost their implementation.

Chapter 3 - The shapes of gold nanoparticles greatly influence their optical and electronic properties. The shape of a gold Nanorods (NRs) is defined by its aspect ratio (AR, length/diameter). Localized surface plasmons (SP) are standing waves of coherent fluctuations of electron charges along the metal-dielectric boundary. As the AR of the gold NRs increases, the energy separation between the two SP resonance peaks increases.

Different wet chemical methods to control the AR of the gold NRs were presented. Better filtration and purification methods were employed to improve the yield of gold NRs. For this centrifugation of low-AR gold NRs were used to separate the spherical nanoparticles from the gold NR solution. A combination of centrifugation and partial dissolution, using a reducing agent for shape separation in the case of high-AR gold NRs were used to improve the overall yield of the NRs in the growth solution.

Various characterization methods i.e., microscopy and spectroscopy post synthesis of gold NRs were presented to determine the AR and LSP resonance. These studies on controllable synthesis and better filtration methods of synthesizing gold NRs into a desired shape, might be useful for application in future nanoelectronics and nanodevices.

Chapter 4 - The current avalanche of research into nanotechnology has led to the examination of various nanoparticle-mediated drug delivery vehicles that can be deployed for diagnostic and therapeutic purposes. Antibody-nanoparticle conjugates can elicit effective and specific targeting at the diseased site, without any significant injury to normal tissues. This review captures an overview of various antibody-nanoparticle conjugating modalities and their potential clinical application. In the midst of the enormous progress made in understanding the molecular and cellular basis of cancer, traditional cancer treatment still remains with both systemic chemotherapy and radiotherapy. Improved formulations in the various protocols of current chemotherapies have the potential to overcome the challenges in the effective

obliteration of the diseased tissue. The application of nanoparticle (NP) drug delivery systems has been studied extensively for a plethora of drugs and drug molecules, with different characteristics such as size, shape, biodegradability, entrapment efficiencies and release profiles. Indeed, nanoparticulate formulations of daunorubicin and doxorubicin are used clinically to treat Kaposi's sarcoma and breast cancer. Despite the successes chalked with NP formulations on drug retention and controlled release, tissue targeting of these formulations could further improve both the therapeutic effectiveness of drugs at the diseased site, whilst reducing collateral damages. Targeting can be achieved through various techniques that complex biomolecules on the surface of the NP to achieve localization and targeting of the drug conjugate. The review first discusses the main classes of NP before shedding light on the current targeting strategies, with particular emphasis on antibodies.

Chapter 5 - In the recent decades, polymeric nanoparticulate drug delivery systems have been recognized due to its unique favorable physicochemical properties such as structurally stable nanoscale size, large surface area, improved solubility of poorly water-soluble drugs, prolonged systemic circulation, sustained and/or controlled drug release, target delivery and combination therapy by delivering multiple therapeutic agents to the same cells. This chapter reviews the numerous methods available to fabricate delivery systems depending upon the physicochemical properties of active ingredients and the polymers. The ideal requirements for designing drug-loaded polymeric nanoparticles are controlled particle size, zeta potential, solubility, and permeation, as well as prolonged circulation, desired release of therapeutically active agents in order to attain the target and specific activity at a predetermined rate and duration. Depending on the method of fabrication, the drug-loaded polymeric nanoparticles could be designed to possess various properties and release characteristics for drug delivery. Upon the fabrication of the drug-loaded polymeric nanoparticles, characterization techniques to determine particle size, zeta potential, drug loading, degradation, drug release kinetics, etc. are discussed. Finally, this chapter reviews recent developments, research investigations as well as clinical applications of the drug-loaded polymeric nanoparticles in optimizing therapeutic outcomes over the recent decades.

Chapter 6 - In this chapter the review on the recent achievements in the field of development of different photoconverters (photodetectors and solar cells) based on nanoheterostructures with quantum dots of germanium on silicon is done. Problems of technology of the creation of such photoconverters by the method of molecular beam epitaxy (MBE) are

considered. Special attention is paid to the questions of optimization of growth conditions in the method of MBE for creation of photodetectors and solar cells with maximum efficiency. Results of investigations of properties of structures with quantum dots and their dependencies on growth parameters, as well as characteristics of quantum dot based photoconverters are presented.

For calculations of dependencies of quantum dots array parameters on synthesis conditions the developed kinetic model of growth of differently shaped quantum dots based on the general nucleation theory is used. This theory is improved by taking into account the change in free energy of island's nucleation due to formation of additional edges of islands and due to dependence of surface energies of facets of quantum dots on the thickness of 2D wetting layer during growth by Stranski–Krastanow mechanism. In the approximation of dependence of surface energy on 2D layer thickness generalized Muller–Kern formula for the equilibrium thickness of the wetting layer is considered.

Results of estimations of noise and signal characteristics of photodetectors with quantum dots of germanium on silicon are presented. Dark current in such detectors caused by thermal emission and tunneling of charge carriers, as well as detectivity in background limited operation mode are calculated. The model of solar cell based on *p-i-n*-structure with Ge quantum dots layers in the intrinsic region is considered. Comparison of estimated characteristics with experimental data on photoconverters is made. Results of investigations of electrophysical parameters of heterostructures with quantum dots by the method of admittance spectroscopy are presented, which confirm the results of theoretical considerations. The obtained results allow us to suppose optimal growth conditions in the method of molecular beam epitaxy for heterostructures with quantum dots of germanium on silicon to create photodetectors with high detectivity and solar cells with improved efficiency.

CONTENTS

In: Advances in Nanotechnology. Volume 17 ISBN: 978-1-53611-004-3
Editors: Z. Bartul and J. Trenor

Chapter 1

ATOMIC NEGATIVE IONS CREATION: APPLICATION IN NANOCATALYSIS

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ABSTRACT

The role of atomic particles and nanoparticles in catalysis continues to attract extensive investigations from both fundamental and industrial perspectives. Recently, we added the novel atomic negative ions to the study of catalysis at the atomic and nano scales. The fundamental mechanism of negative-ion catalysis is the weakening or breaking up of moleular bonds in the transition state by the negative ion. Here the complex angular momentum method is used to generate atomic negative ions from selected complex atoms through the dramatically sharp resonances, manifesting electron attachment, that characterize the calculated low-energy electron elastic total cross sections (TCSs). The atomic Au TCSs are used as the benchmark for selecting the appropriate atoms. Ramsauer–Townsend (R-T) minima, shape resonances (SRs) and binding energies (BEs) of the resultant anions formed during the electron-atom collisions are extracted. The striking features of these TCSs, exploited in this chapter, are the appearance of a dramatically sharp resonance with large BE in the ground state second R-T minimum and a broad deep R-T minimum in the excited state TCS; thus providing an

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excellent environment and mechanism for breaking up molecular bonds in new molecules creation from atoms as well as in anionic nanocatalysis. Selected generated negative ions are demonstrated in various applications, including in the dynamic negative-ion nanocatalysis of various chemical reactions: 1) Catalysis of water to peroxide, 2) Negative-ion catalysis of light, intermediate, and heavy water to corresponding peroxides, 3) Methane oxidation to methanol without CO_2 emission, 4) Multiple atom super nanocatalysts, 5) Single atom multifunctional nanocatalysts and sensors, 6) Tunable catalysis of water to peroxide with atomic catalysts and 7) Novel mechanism for creating long-lived (metastable) atomic negative ions. We conclude by discussing prospects of using some of the gaseous multiply functionalized anionic nanocatalysts to resolve the problem of deep oxidation in the partial oxidation of methane to methanol.

Keywords: nanocatalysis, atomic negative ions, resonances, elastic cross sections, complex angular momentum, binding energies

1. INTRODUCTION

The importance and utility of negative ions in terrestrial and stellar atmospheres as well as in industrially important plasmas for device fabrication are well documented [1-5]. Low energy collisions, resulting in negative ion formation as intermediate resonances, provide a special insight into quantum dynamics [6]. Consequently, low energy electron collisions necessitate the careful determination of binding energies (BEs), electron affinities (EAs), Ramsauer-Townsend (R-T) minima and shape resonances (SRs). The R-T effect has been used to understand sympathetic cooling and to produce cold molecules using natural fermions [7], since at the R-T minimum the electrons pass through the molecules as though they were transparent. Accurate atomic and molecular affinities are essential for the understanding of chemical reactions involving negative ions [5].

Recently, Freakley et al. [8] reported a significant improvement in the direct synthesis of H_2O_2 over using a Pd-Sn alloy. Previously, the direct synthesis of hydrogen peroxide from H_2 and O_2 using supported Au, Pd and Au-Pd nanoparticle catalysts was reported [9, 10]. The experiments also found that the addition of Pd to the Au catalyst increased the rate of H_2O_2 synthesis significantly as well as the concentration of the H_2O_2 formed. The anionic complexes $Au^-(H_2O)_1$ and $Au^-(H_2O)_2$ have been characterized as atomic gold anion interacting with one and two water molecules, respectively, i.e., as