

B Nanomaterials for Bioimaging and Biosensing



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Symposium B

Nanomaterials for Bioimaging and Biosensing

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Scope of Symposium

Advanced nanomaterials display unique robustness, physical and chemical properties. They offer a myriad of opportunities to couple analyte-receptor interactions into observable and transduced signals. They also provide unique opportunities for bioimaging applications. In this symposium, different aspects and types of nanomaterials for bioimaging and biosensing will be covered. The objective is to draw a diverse group of researchers including chemists, material scientists, physicists, engineers, medical doctors, and biochemists to discuss new ideas, experimental findings, prototypes, and challenges ranging from synthesis, and characterization to the application of nanomaterials for bioimaging and biosensing.

Symposium Topics

Nanomaterials for Bioimaging

- Quantum Dots – Synthesis, Properties, and Surface Functionalization
- Metallic and Magnetic Nanoparticles – Synthesis, Properties, and Surface Functionalization
- Synthesis, Shape Control and Assembly of Nanorods, Nanotubes, and Nanowires
- Composites of Heterogeneous Nanostructures
- Nanomaterials for Targeting and Labeling
- Magnetic Nanoparticles for Bioimaging, Bioseparation and Drug Targeting
- Multifunctional Nanoparticles for Biolabeling, Bioseparation and Bioimaging

Molecular design and synthesis of nanomaterials for sensing

- Bioconjugate Chemistry to Link Nanomaterials with Biomolecules
- Signal Transducing Mechanisms between Analyte/Receptor and Nanomaterials
- Nanoparticles, Nanorods, and Nanowires for Molecular and Biological sensing
- Nanostuctured Polymers and Polymer Thin Films for Biosensing
- Nanomaterials-Based Electronic Devices for Molecular and Biological Sensing
- Integration of Nanomaterials to Diagnostic and Bioinformatic systems

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Abstracts

A00006-00018

Examination of Au Nanocrystal-Directed Growth of Au-Cu₂O Core-Shell Nanostructures

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Cuprous oxide (Cu₂O) is a *p*-type semiconductor with a direct band gap of 2.17 eV. It has been demonstrated to be useful for applications in gas sensing, CO oxidation, photocatalysis, photoactivated splitting of water, and organic synthesis. We have developed simple aqueous solution approaches for the fabrication of Cu₂O nanocrystals and completed three studies. They involve the synthesis of truncated Cu₂O nanocubes with sizes ranging from 40 to 420 nm, the synthesis of Cu₂O nanocrystals with systematic shape evolution from cubic to octahedral structures, and the fabrication of Cu₂O nanocages and nanoframes by particle aggregation and acidic etching.

Recently, we have found a facile Au nanocrystal-directed growth strategy for the synthesis of Au-Cu₂O core-shell heterostructures with complex but symmetrical morphologies. Octahedral Au nanocrystals, Au nanoplates, Au nanorods, etc. have been used to serve as the cores. For instance, by using high-aspect-ratio Au nanorods as the cores, symmetrical star fruit-shaped Cu₂O shells were formed. With octahedral Au serving as the cores, truncated octahedral shells were produced. By examining the intermediate products generated at different growth stages, we found that the preformed shells would enclose these Au cores before the final formation of the complete core-shell structures. This observation indicates that the gold cores can direct the morphological development of the Cu₂O shells, unlike the conventional shell formation mechanism in which conformal growth is typically observed.

A00023-00024

Facile Synthesis of Cu₂O Nanocrystals

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We will first present a facile method for the synthesis of cuprous oxide nanocrystals with systematic shape evolution. Monodisperse truncated cubic, cuboctahedral, truncated octahedral, and octahedral nanocrystals can be synthesized directly in an aqueous solution of CuCl₂, sodium dodecyl sulfate (SDS) surfactant, hydroxylamine (NH₂OH·HCl) reductant, and NaOH by simply varying the volume of hydroxylamine added to the reaction mixture.

Adjustment of the volume of NaOH added provides a means to vary the particle size. All four samples displayed activity toward photodegradation of Rhodamine B molecules, but truncated octahedral and octahedral nanocrystals exhibited a higher extent of the photodecomposition reaction, suggesting the {111} faces of Cu₂O nanostructures are catalytically more active than the {100} faces.

Next we will describe a simple approach for the fabrication of Cu₂O nanocages and nanoframes possessing an unusual truncated rhombic dodecahedral structure. An aqueous solution containing CuCl₂, SDS surfactant, NH₂OH·HCl reductant, HCl, and NaOH was prepared, with the reagents introduced in the order listed. Rapid seed-particle aggregation and surface reconstruction of the intermediate structure resulted in the growth of type-I nanoframes, which have only {110} skeleton faces and empty {100} faces, 45 min after mixing the reagents. Continued crystal growth for additional 75 min produced nanocages with filled {100} faces. Selective acidic etching over the {110} faces of the nanocages by HCl via the addition of ethanol followed by sonication of the solution led to the formation of type-II nanoframes, which have elliptical pores on the {110} faces. Without addition of ethanol, random etching of the nanocages can occur at a slow rate. Octahedral gold nanocrystals and high-aspect-ratio gold nanorods were successfully encapsulated in the interiors of these Cu₂O nanocages by adding the gold nanostructures into the reaction solution.

A00072-04679

Synthesis, Structure, Electrochemical Properties of Nano Layered Organic-inorganic Perovskites Containing Fe(CN)₆³⁻ Layers and its Application for Detection of DNA Hybridization

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The layered organic-inorganic perovskites are a subclass of the organic-inorganic hybrids and have recently attracted substantial interest due to their unique electrical, magnetic, and optical properties. The simplest and most common examples of layered organic-inorganic perovskite hybrids have the general formula (R-NH₃)₂MX₄ (M = divalent metal, X = halogen). There are a few layered organic-inorganic perovskites containing trivalent metal. Conventional perovskite frameworks, trivalent metal ions halide lattices generally are composed of distorted MX₆ (X = halogen) octahedra. These MX₆ octahedra form discrete (i.e. mononuclear) or extended (i.e. polynuclear) inorganic networks of corner-, edge-, or face-sharing

octahedra, leading to an extensive family of metal (III) halogenoanions (e.g., MX_4^- , MX_5^{2-} , MX_6^{3-} , $\text{M}_2\text{X}_9^{3-}$, $\text{M}_2\text{X}_{11}^{5-}$, $\text{M}_3\text{X}_{12}^{3-}$, $\text{M}_4\text{X}_{18}^{6-}$, $\text{M}_5\text{X}_{18}^{3-}$, $\text{M}_6\text{X}_{22}^{4-}$, and $\text{M}_8\text{X}_{30}^{6-}$). Within these networks, the inorganic sections of perovskite hybrids incline to diversification. In this study, halogen was replaced by CN⁻ to form stable inorganic sheets --- $\text{Fe}(\text{CN})_6^{3-}$ layer, which can avoid MX_6 octahedra form discrete or extended inorganic networks. Steady nano organic-inorganic perovskite hybrids with $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$, $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$ and $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$ (AMP = aminomethylpyridine) were formed in the air. Each structure shows an unusual layered organic-inorganic structural type. The hybrids enveloped in paraffin, respectively, to prepare hybrid paste electrode (HPE) (HPE-2 with $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$, HPE-3 with $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$, HPE-4 with $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$). Three hybrids in HPEs showed good electrochemical characteristics. The sequence of redox activity is $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6 > [\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6 > [\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$ and that of electrocatalytic characteristics is $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6 > [\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6 > [\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$. Three hybrids have been employed to investigate the interaction between DNA and three hybrids. The results indicate that between $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$ and DNA is useful and $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$ can detect the hybridization of DNA. And the interaction between $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$ and DNA and between $[\text{H}_2\text{-AMP}]_{3/2}\text{Fe}(\text{CN})_6$ and DNA is slender. Three HPEs display remarkable electrochemical sensitivity and stability. The variation coefficients (RSD) of repeatedly successive and interval assays are less than 2.5%. The chemical and physical stability of three hybrids is satisfactory.

A00178-00567

Synthesis and Characterisation of Magnetic-Fluorescent Composite Colloidal Nanostructures

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The application fields of optical or magnetic inorganic nanoparticles are continuously increasing. Inorganic luminescent materials have practical applications in a variety of devices from conventional TV to electroluminescent, plasma or field emission displays and nowadays luminescent labelling is currently important bio-techniques. Magnetic nanoparticles are of great interest for researchers from a wide range of disciplines, including magnetic fluids, catalysis, biotechnology/biomedicine, magnetic resonance imaging, data storage, and environmental remediation.

Recently, nanocomposite materials provide the possibility for enhanced multifunctional properties. Specially, nanometer sized particles, such as quantum dot (QDs) or superparamagnetic oxide have novel optical, electronic and magnetic properties that are not available for either molecules or bulk counter part solids. One advantage is that this type of bifunctional nanoparticles can be manipulated with magnets and detected by fluorescence.

In this communication, we report fluorescent, photostable and magnetic nanoparticles in the nanoscale range. The synthesis and characterisation of various colloidal nanostructures as $\text{M}@\text{SiO}_2$ ($\text{M} = \gamma\text{-Fe}_2\text{O}_3$, ZnFe_2O_4 and/or $\text{Cs}_2\text{Mo}_6\text{Br}_{14}$) or $\gamma\text{-Fe}_2\text{O}_3$ -QDs will be presented.

At the outset, the synthesis of nanoparticles under restricted environments offered by water-in-oil microemulsions provides excellent control over particle size and shape and interparticle spacing. These environments have been used in the synthesis of bi-functional silica nanoparticles with a magnetic and optical core (average diameter : 50 nm). Secondly, we will describe the preparation of composite magnetic QD bioconjugable micelles by simultaneous encapsulation of hydrophobic CdSe/ZnS QDs (2-4 nm) and magnetic $\gamma\text{-Fe}_2\text{O}_3$ nanoparticles (3-4 nm) into micelles of synthetic functional PEG amphiphiles bearing a bioactivable terminal group.

A00222-00418

Assembly of Gold Nanostructures into Chains and Three-Dimensional, Highly Ordered Superstructures

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Gold nanostructures exhibit rich optical properties that result from the localized surface plasmon resonance. A number of attractive applications have been demonstrated with discrete gold nanostructures, including optical data storage, metallic barcodes, sensing, bioimaging, controlled drug delivery and photothermal therapy. Future scientific and technological applications of gold nanostructures require the capability to assemble into complex one-, two-, or even three-dimensional functional architectures. We have developed methods for both the pH-controlled, reversible assembly and disassembly of gold nanorods in solutions (Chem. Commun. 2007, 1816; Small 2008, 4, 1287) and the formation of three-dimensional, highly ordered assemblies of gold nanostructures with varying sizes and shapes (Angew. Chem. Int. Ed. 2008, 47, 9685).

In the pH-controlled assembly of gold nanorods, small thiol molecules, including 3-mercaptopropionic acid, 11-mercaptopundecanoic acid, cysteine, and glutathione, have been employed as assembling agents. Depending on

the concentration and molecular size, they can preferentially bind to either the ends or side surfaces of gold nanorods. The binding of the thiol molecules to the ends induces the end-to-end assembly of gold nanorods, and their binding to the side surfaces induces the side-by-side assembly of gold nanorods. Moreover, the assembly and disassembly in both manners can be controlled by pH. They can be cycled many times by adjusting the solution pH to appropriate values. Finite-difference time-domain calculations have further been performed on assembled gold nanorods at varying gap distances. The calculated relationship between the plasmon shift and nanorod spacing has been used to estimate the spacing between assembled nanorods.

A simple droplet evaporation method has been employed to assemble variously-shaped gold nanostructures into three-dimensional, highly ordered superstructures. The resultant assemblies are strongly dependent on the shape of gold nanostructures for single-component systems. Nanorods, nanopolyhedra, nanocubes, and nanobipyramids are found to form nematic/smectic, hexagonally packed, tetragonally packed, and nematic/three-dimensional ordered superstructures, respectively. Binary mixtures of gold nanorods of different sizes either assemble into nematic superstructures or exhibit self-separation, with each nanorod sample forming its own smectic superstructure. The assembly behavior of binary nanorod mixtures is observed to be dependent on the relative diameters of two nanorod samples. Moreover, binary mixtures of gold nanorods and nanopolyhedra assemble into an alternating ordered superstructure. Two photon-excited photoluminescence measurements show that the photoluminescence intensity of highly ordered gold nanorod superstructures is enhanced in comparison to that of disordered nanorod assemblies. We believe that our assembly technique can be extended to other anisotropic nanostructured materials for the fabrication of large-scale ordered superstructures with interesting physical properties.

A00278-00509

Large-scale Preparation of Semiconductor Photoluminescent Quantum Dots and Their Bioimaging Application

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Feng LONG; Lin-Jing SHEN

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Photoluminescent semiconductor nanocrystals or quantum dots (QDs) are of great fundamental and industrial interest because of their size-dependent photoluminescent properties. Generally, QDs are prepared on a small scale by utilizing very expensive ligands and solvents. However, the complicated combination from different ligand systems often results in the poor reproducibility of QD preparation.

In addition, these methods indicate some limitations in process manipulation and material cost. Therefore, the facile large-scale preparation of low-cost QDs with commercial potential is of critical importance for many purposes. This initiated broad exploration on the large-scale preparation of low-cost QDs through employing cheap, environmentally benign raw materials and relatively lower reaction temperatures. Here we introduce several simple methods to low-cost large-scale preparation of CdSe, CdTe etc. as well as their bioimaging applications.

A00278-00510

A Versatile Energy-Efficient Approach to CdTe Quantum Dots at Moderate Temperature

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We introduce here a facile preparation of photoluminescent semiconductor CdTe nanocrystals or quantum dots (QDs) at moderate temperature. The reaction was conducted in the presence of aliphatic acid at relatively low temperature between 80-140°C, which is much lower than high temperature in conventional organic-phase preparation of CdTe QD. Considering the pressure of energy cost and environmental pollution, this lower-temperature route to synthesize CdTe semiconductor quantum dots provides a promising alternative to current mainstreaming method of CdTe QDs.

A00304-00562

Self-assembling Optoelectronic Nanostructures Inspired by β -amyloids: Synthesis, Characterization and Prospects for Bioelectronics

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Fields as seemingly disparate as photovoltaics and tissue engineering have witnessed exciting advances based upon biomimetic design. While humanity has become quite proficient in the construction of elegant and complex molecular structures, our ability to control supramolecular interactions within functional nanostructures remains in its infancy. In this contribution, I will highlight an approach from our laboratory to exert supramolecular control over the orientation and ordering of synthetic organic electronic moieties in aqueous media. We developed π -conjugated "amino acids" that allow us to incorporate electronic functionality directly into the backbones of oligopeptides.

The peptide segments may be varied to control assembly or control the presentation of biological signals, and specific environmental stimuli can be envisioned to trigger the self-assembly process. The self-assembly process leads to the formation of self-supporting hydrogels, and visualization of these materials via AFM reveals the presence of well-defined one-dimensional aggregates with features smaller than 10 nm. We will discuss spectroscopic measurements employed to further characterize the assembly process, and prospects for biomaterials applications that can exploit the semiconductive nature of these nanostructures will be addressed.

A00326-00602

Origin of Manganese Luminescence Enhancement in Mn-doped ZnS Nanoparticles

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Doping ZnS nanocrystals with Mn²⁺ ions allows them to emit in the orange region and opens up its potential for applications in electroluminescent displays, spintronics and biomedical labelling. In this study, we focused on undoped ZnS and Mn-doped ZnS nanoparticles that are fabricated using a single molecule precursor method. The ZnS nanocrystals were grown from the thermal decomposition of 2,2'-bipyridine(Zn(SC{O}Ph)₂) in an alkylamine medium under a nitrogen atmosphere. The Mn-doped ZnS nanocrystals were grown by injecting a solution containing MnCl₂·4H₂O, 2,2'-bipyridine(Zn(SC{O}Ph)₂) precursor, trioctylphosphine and oleylamine into a hot solution (280°C) of hexadecylamine and 1-octadecene. Nanoparticles with good crystallinity and small size distribution can be obtained using this method. Their Zn blende crystal structure was verified using high resolution transmission electron microscopy and x-ray diffraction. Extended X-ray Absorption Fine Structure confirmed the occupation of substitutional Zn²⁺ sites by the Mn²⁺ dopants. Our recent low temperature photoluminescence measurements have revealed phonons play a dominant role in the energy transfer between the ZnS host and the Mn subsystem. Time-resolved photoluminescence studies involving Mn:ZnS/ZnS core-shell samples have also revealed that the site dependence on the Mn luminescence lifetimes. Further studies involving the overcoating of the Mn:ZnS studies with pyridine shows that the Mn luminescence lifetimes are also modified. Mn:ZnS nanoparticles with enhanced luminescence yield for bio-imaging applications could be fabricated through careful tuning of the particle size and surface functionalization.

A00370-01756

Development of Nanoarchitected Amperometric Biosensors

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Transition metal nanoparticles have gained enormous interest in the various areas of science and technology owing to their excellent optical, electronic and catalytic properties. The properties of these metal particles strongly depend on their shape, size and morphology and are very attractive for practical applications, particularly in the development of nanoscale devices. Au and Pt nanoparticles received significant interest in the development of biosensing devices. The large surface-to-volume ratio, biocompatibility and the fascinating optical and catalytic properties made them an ideal candidate in the development of biosensors. The integration of nanoparticles and biocatalyst with the transducer is a promising approach in the development of highly sensitive biosensing platform. Our group is interested in exploring the electrocatalytic and bioelectrocatalytic properties of nanosized metal particles. Herein we describe the electrocatalytic activity of nanosized Pt particles and the development of amperometric biosensing platform for the sensing of bioanalytes. We have developed a highly sensitive biosensor for clinically important analytes such as glucose, uric acid etc. by integrating the corresponding oxidase enzymes and Pt nanoparticles with the transducer.

Spherical shape Pt nanoparticles were synthesized by wet chemical route and were immobilized on a conducting support functionalized with 3-D silicate network. The nanoparticles were characterized by FESEM, XRD, XPS and electrochemical techniques. The particles on the conducting support have an average size of 9 nm. The electrocatalytic activity of the nanoparticles towards hydrogen peroxide was examined. The nanoparticles on the conducting support efficiently catalyze the oxidation of hydrogen peroxide at less positive potential. Large decrease in the overpotential and enhancement in the peak current with respect to the bulk Pt electrode were noticed. The nanoparticle based transducer is highly sensitive and could detect hydrogen peroxide at the sub-nanomolar level without any redox mediator or enzyme. Particle coverage controls the catalytic activity.

Nanoarchitected biosensor was developed by integrating the oxidase enzymes and the nanoparticle with the transducer. The enzyme on the conducting support retains its activity and efficiently catalyze the oxidation of substrate. Enzymatically generated hydrogen peroxide was successfully detected by the nanoparticle on the biosensor. Amperometric sensing of clinically important bioanalytes such as glucose, uric acid etc. has been successfully achieved. The analytical application of the

nanoarchitected biosensor in the clinical analysis was examined with serum samples. The concentration of bioanalyte in the real samples obtained with the sensor is closely similar to those of the clinical laboratory. High sensitivity, excellent reproducibility and long term stability were noticed. The details will be presented.

This work was financially supported by Department of Science and Technology, New Delhi, India

A00384-00705

Conjugated Polyelectrolytes as Platforms for Biomacromolecule Detections

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Recently, conjugated polyelectrolytes have provided a unique platform for chemical and biological sensors in view of their optical signal amplification effect. We and others have utilized this optical property of conjugated polyelectrolytes to detect DNA, RNA, protein and metal ions with high sensitivity. Our recent studies showed that the conjugated polyelectrolytes can be used for detecting genetic disorders, such as single nucleotide polymorphisms (SNPs) and DNA methylation, that can cause cancer. SNPs are the most frequent form of sequence variation in the human genome, which are useful for identifying the disease-causing genes. Genotyping large number of SNPs will take a deep insight into understanding and clinically diagnosing the complex diseases. DNA methyltransferases catalyze the covalent addition of methyl group to adenine and cytosine residues in DNA. The resulting DNA methylation plays a key role in control of gene expression, genomic integrity maintenance and cancer origin. We use conjugated polyelectrolytes/DNA complexes combining with fluorescence resonance energy transfer (FRET) processes to assay SNP genotyping and DNA methylation, thus offering new assay strategies based on conjugated polyelectrolytes.

We also showed that the complexes of conjugated polyelectrolytes/enzymatic substrates by electrostatic interactions can be utilized as probes for continuous and sensitive fluorescence assays for disease-related enzymes, such as nuclease, phosphatase, protease, acetylcholinesterase and kinase. The technique also provides a promising application in drug screening based on the inhibition of the cleavage reactions.

A00426-01578

Synthesis and Optical Spectroscopy of CdSe Quantum Dots Semiconductor

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High optical quality CdSe quantum dots with average size at the new Bohr radius was synthesized using non-TOP approach. The as-synthesized CdSe QDs were characterized by XRD, TEM and UV-vis spectrophotometer. The XRD pattern showed zinc blend phase of the CdSe QDs. The XRD and TEM results showed that crystallite sizes and physical sizes of CdSe QDs were less than 10 nm. The non-TOP based route enables us to achieve high quality of CdSe QDs with sharp ultraviolet and visible absorption peaks with controllable sizes (2-5 nm). The CdSe quantum dots exhibited strong blue shift and discrete energy states which were significantly modified from bulk crystals. Atomic-like discrete energy states of exciton indicate a quantum confinement effect. The effective band gap of CdSe quantum dots calculated (1.96-2.59 eV) show increment value as the reaction time increases.

We are grateful for the support from the University of Malaya PPP fund (No. PS079/2008C).

A00471-01294

Advanced Functional Nanocrystals for Bioimaging and Biosensing

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Colloidal semiconductor nanocrystals (quantum dots) have attracted great attention for their distinguished roles in fundamental studies and technical applications such as biological labeling and optoelectronic devices. In the last two decades, the main efforts have been focused on the preparation of size-tunable binary or core-shell nanocrystals with different emission colors. In our research, we also focus on the development of highly luminescent composition-tunable quantum dots across the whole visible spectrum. The successful preparation of high-quality composition-tunable quantum dots makes the new class of nanomaterials very promising as multicolor biological nanoprobe for imaging, sensing, and drug delivery applications. There is an emphasis on the development of ultrasensitive imaging/spectroscopic detection for multiplexed analysis at cellular or tissue levels. Quantitative multi-parameter analysis of multianalytes is being carried out, which could provide a direct way to identify sets of genes and proteins correlating with certain diseases.

A00491-00901

Controlling the Optical Properties of Inorganic Nanocrystal HeterostructuresGregory SCHOLES*Department of Chemistry, University of Toronto, Toronto ON, Canada*

The sophistication with which we can now prepare and characterize inorganic nanoparticles has inspired new areas of research into the fundamental properties and applications of these fascinating nanoscale systems. In this talk some of the recent ideas concerning control of their optical properties will be discussed, focusing on semiconductor nanocrystals. A particular emphasis of the talk will be the control of optical properties in nanocrystal heterostructures. As one example, the ultrafast dynamics of charge separation and concomitant spin relaxation in CdSe–CdTe ‘donor-acceptor’ nanorods will be reported and compared to various series of core-shell type-II heterostructures. It will be shown how electron-hole separation can be employed to decrease electron spin relaxation times by an order of magnitude. In a second example, it will be reported how the size and shape of a seed crystal can be used to direct the shape of heterostructure growth.

A00497-00913

The Preparation of Colloidally Stable, Water Soluble, Biocompatible, Semiconductor Nanocrystals with a Small Hydrodynamic DiameterPaul MULVANEY*School of Chemistry & Bio21 Institute, University of Melbourne, VIC, Australia*

We report a simple, economical method for generating water soluble, biocompatible nanocrystals that are colloidally robust and have a small hydrodynamic diameter. The nanocrystal phase transfer technique utilizes a low molecular weight amphiphilic polymer that is formed via maleic anhydride coupling of poly(styrene-co-maleic anhydride) with either ethanolamine or Jeffamine M-1000 polyetheramine. The polymer encapsulated water soluble nanocrystals exhibit the same optical spectra as those formed initially in organic solvents, preserve photoluminescence intensities, are colloidally stable over a wide pH range (pH 3–13), have a small hydrodynamic diameter and exhibit low levels of non-specific binding to cells.

A00519-00948

Optical Properties of Manganese-Doped ZnSe Nanoparticles Synthesized by Wet Chemical RouteSee Yau LEE¹; Sabar Derita HUTAGALUNG¹; Azizan AZIZ¹; Ishak MAT²*1. School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia, Penang, Malaysia**2. Advanced Medical and Dental Institute, Universiti Sains Malaysia, Penang, Malaysia*

Monodispersed zinc selenide (ZnSe) nanoparticles were synthesized via wet chemical route. Zinc chloride (ZnCl₂) and sodium hydrogen selenide (NaHSe) were used as the source materials to synthesize ZnSe nanoparticles. Briefly, ZnCl₂ and NaHSe solution were prepared at same concentration of 50 mM and mixed together to obtain stoichiometric composition ZnSe. In order to enhance photoluminescence properties, manganese dopant with vary concentration was added into the ZnSe nanoparticles. The structural, morphological and luminescence properties of the undoped and Mn-doped ZnSe nanoparticles were investigated using X-ray diffraction, scanning electron microscopy, UV-Vis spectrometer and transmission electron microscopy. Results shown the Mn-doped ZnSe nanoparticles reveal higher photoluminescence properties than undoped ZnSe nanoparticles. The highly luminescent and photostable Mn-doped ZnSe nanocrystals can further be used as the luminescent biomarkers.

A00519-00952

Effect of Surfactant Concentration on the Formation of ZnSe Nanoparticles Synthesized by Wet Chemical RouteSee Yau LEE¹; Sabar Derita HUTAGALUNG¹; Azizan AZIZ¹; Ishak MAT²*1. School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia, Penang, Malaysia**2. Advanced Medical and Dental Institute, Universiti Sains Malaysia, Penang, Malaysia*

Monodispersed zinc selenide (ZnSe) nanoparticles were synthesized via wet chemical route. Zinc chloride (ZnCl₂) and sodium hydrogen selenide (NaHSe) were used as the source materials to synthesize ZnSe nanoparticles. Briefly, ZnCl₂ and NaHSe solution were prepared at same concentration of 50 mM and mixed together to obtain stoichiometric composition ZnSe. Polyvinyl alcohol (PVA) was added to the mixed solution as a stabilizer. In order to investigate the effect of surfactant concentration on the formation of ZnSe nanoparticles, the concentration of PVA were varied. Results shown that the increase of the PVA concentration tend to decrease the ZnSe nanoparticle size.

A00548-01009**Oil-in-water Emulsion Synthesis of Hydroxyapatite with Controlled Size and Homogeneous Morphology**

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College of Materials Science and Engineering, South China University of Technology, China

In this paper, we prepared different morphology of nano-hydroxyapatite particles by oil-in-water emulsion synthesis process, such as nano-sphere, nano-needle, nano-wire, nano-rod and nano-belt. The samples were characterized by TEM, HRTEM, XRD, FT-IR and UV-vis spectra. The results showed that the nano-wires consisted uniform nano-fibers with diameter of 1.6 nm and length of 250 nm. The sphere particle size was 55 ~ 60 nm. According to the UV-vis spectra analysis, the energy bands for the nano-hydroxyapatite were 1.60 eV, 1.64 eV, and 1.69 eV, corresponding to nano-rod, nano-sphere and nanowire, respectively.

A00582-02818**Preparations and Evaluations of Magnetically Guided Nanoparticles Carrying Thrombolytic Drug**Jyh-Ping CHEN¹; Yunn-Hwa MA²; Pei-Ching YANG¹; Show-Yunn WU²*1. Department of Chemical and Materials Engineering, Chang Gung University, Taiwan**2. Department of Physiology and Pharmacology, Chang Gung University, Taiwan*

Recombinant tissue plasminogen activator (rtPA) has been used for thromboembolic diseases with hemorrhagic side effects. In this study, we investigated the feasibility and efficacy of target thrombolysis with rtPA covalently bound to magnetic nanoparticles (MNP). Polyacrylic acid-coated magnetic nanoparticles (PAA-MNP) were synthesized and characterized. rtPA was immobilized to PAA-MNP through carbodiimide-mediated amide bond formation between the amino groups of rtPA and the carboxyl groups of PAA-MNP. The enzyme activities of the bound rtPA, as measured by a chromogenic substrate assay and ¹²⁵I-fibrinolysis assay, were 87 ± 1% and 86 ± 3% of that of the free rtPA, and improved storage stability at 4°C was observed. Under magnetic guidance, the thrombolytic activity of PAA-MNP-rtPA, at a dose equivalent to 0.2 mg/kg of rtPA, was determined by ultrasound and laser Doppler flowmetry in a rat embolic model. Intra-arterial administration of PAA-MNP-rtPA reversed the iliac blood flow to 82% of that before the clot lodging within 75 min, whereas the PAA-MNP or equivalent amount of free rtPA exerted no improvement on hemodynamics. At the end of 2 hr periods, PAA-MNP-rtPA did not alter levels of hemoglobin or hematocrit, or counts of red blood cells,

white blood cells or platelets. Immobilization of rtPA to PAA-MNP with covalent binding resulted in a stable drug preparation, consistent drug concentration around the target site, and subsequently reproducible and effective target thrombolysis in the rat embolic model. With this novel approach, 20% rtPA may achieve equivalent thrombolysis/hemodynamic recovery induced by a regular dose of the drug.

A00588-02151**Synthesis and Characterization of Nano-Crystalline LaF₃**Arun Kumar DORAI^{1,2}; Selvasekarapandian SUBRAMANIAM³; Hema MUTHUSAMY^{1,2}; Nithya HELLER²; Sakunthala AYYASAMY²*1. Department of Physics, Bharathiar University, Tamilnadu, India**2. Physics Division, DRDO-BU Centre for Life Sciences, Tamilnadu, India**3. Kalasalingam University, Tamilnadu, India*

Electrochemical sensors have been widely used in applications such as critical care, safety, industrial hygiene, process controls, emissions monitoring etc. Solid electrolyte sensors are good electrolytes and offer excellent potential for use in electrochemical sensors. Fluorides have been widely applied in many fields such as sensors, batteries and actuators, and additive and wear resistant materials. Fluoride ion is one of the smallest anion with high mobility, exhibit high ionic conductivity at ambient temperatures. As the world approaches towards nanotechnology, it is an effective method to increase the ionic conductivity of fluorides by having them in nano-size. As the particle size approaches the nano scale the ionic conductivity increases by an order of magnitude. Lanthanum Fluoride is an excellent fluoride ion conductor among other rare earth fluorides. Lanthanum Fluoride based chemical sensors have been applied for sensing the fluoride, oxygen and carbon monoxide because of its high chemical stability and high ionic conductivity. Nowadays many techniques has been employed to synthesize nano crystalline Lanthanum Fluoride (LaF₃). Nano crystalline Lanthanum Fluoride has been synthesized by simple method of direct precipitation from aqueous solution. Particle size and structure of LaF₃ nano-crystalline powder has been analyzed by XRD and SEM. From the XRD Spectra the particle size has been found to be in the range 10 -20 nm. The ionic conductivity of nano crystalline LaF₃ bulk material has been studied using AC impedance spectroscopy in the frequency range 42 Hz to 1 MHz. The ionic conductivity of nano crystalline LaF₃ bulk material is found to be higher than that of the single crystal LaF₃. Dielectric spectra and Modulus spectra analysis has been carried out using AC impedance technique.

A00643-04943

Nano-scale Self-Assembly for Functional Materials and DevicesJatinderVir YAKHMI*Physics Group, Bhabha Atomic Research Centre, Mumbai, India*

In the present talk, we highlight our attempts to utilize self-assembly methods to prepare functional molecular materials (even single crystals) and their films (SAMs, LB or electrochemically grown) to fabricate devices. We shall describe the use of DNA, or a SAM, or simply a surfactant monolayer at the air-water-interface, as templates for growth of crystals of functional materials. We shall discuss our attempts to prepare gas sensors from LB films of polymers, or K-ion sensors from LB films of surfactant encapsulated crystals of Prussian Blue Analogues. We discuss magnetic pole reversal and bipolar switching in a Prussian blue type molecular magnet $\text{Cu}_{0.73}\text{Mn}_{0.77}[\text{Fe}(\text{CN})_6]_z\text{H}_2\text{O}$, as well as magneto-caloric effect in it.

We briefly describe efforts in our lab to fabricate efficient gas sensors from nano-phase materials, viz. highly sensitive H_2S gas sensor based on CuO doped SnO_2 nanowires, Cl_2 -gas sensors using Te-nanotubes or from ZnO-nanowires modified polypyrrole films, H_2S gas sensors using single crystal In_2O_3 whiskers, etc.

Long alkyl chains consisting of σ bonds can act as dielectric, σ - π molecules as rectifier and/or transistor and σ - π - σ molecules as resonant tunnel diodes. We shall describe our efforts to fabricate molecule/Si hybrid molecular electronic components, using self-assembled mono/multilayers on Si. In particular, we shall touch upon electrografting of organic molecules on Si, design of new porphyrin-based σ - π molecules, and reduction of the dielectric layer thickness. We shall conclude by describing the fabrication of phthalocyanine-fullerene photovoltaic cells, in our labs.

A00664-01254

Conductive Molecular and Nanostructural Building Blocks for DNA BiosensingHsiao-hua YU*Yu Initiative Research Unit, RIKEN Advanced Science Institute, Wako, Japan*

Electrical conducting polymers (ECPs) have shown promise in biosensor applications. A key advantage of ECP-based sensors is the amplified sensitivity to the existence of small amount of analytes. Among them, poly(3,4-ethylenedioxythiophene)s (PEDOTs) are of interest due to its long-term stability. PEDOT has shown great stability in aqueous solution and compatibility with biofluids. Three different approaches involving PEDOT

nanobiointerfaces, EDOT intercalators, and label-free biomolecules detection from nanostructured PEDOT are currently being explored. We assembled functionalized PEDOT with different nanostructures to increase the sensitivity of the material. Combining with proper device fabrication and system integration, PEDOTs provide an ideal platform as fast, sensitive, and automated biosensors for biological applications.

A00720-01285

Multicolor Tuning of Lanthanide-doped Down- and Up-conversion Nanocrystals by Single Wavelength ExcitationFeng WANG; Xuejia XUE; Xiaogang LIU*Department of Chemistry, National University of Singapore, Singapore*

Fluorescent labeling with multicolor probes has applications in areas as diverse as drug discovery, DNA sequencing, fluorescent microscopy, and clinical diagnostics. Most common dye probes have low photobleaching thresholds, require different excitation wavelengths, and exhibit broad emission spectra. In contrast, quantum dots (QDs) exhibit relatively narrow emission band widths of 20 to 30 nm (full width at half maximum (FWHM)) under single-wavelength excitation, but they can suffer from cytotoxicity in vivo, fluorescence intermittency, and limited distinguishable features in the emission spectra. As an alternative to dye molecules and QDs, lanthanide-doped nanocrystals have been suggested as a promising new class of fluorescent probes. They show superior chemical and optical properties, including low toxicity, large effective Stokes shifts, sharp emission band widths of 10 to 20 nm (FWHM), as well as high resistance to photobleaching, blinking, and photochemical degradation. More importantly, Ln-doped nanocrystals generally show a distinct set of sharp emission peaks arising from f-f orbital electronic transitions, which should provide spectroscopic fingerprints for accurate interpretation in the event of overlapping emission spectra. These unique properties, coupled with their size- and shape-independent luminescent phenomena should make lanthanide-doped nanocrystals particularly suitable as fluorescent probes for multicolor labeling applications. However, despite the significant efforts on the synthesis of water soluble lanthanide-doped nanocrystals for use as fluorescent probes, preparation of multicolor nanocrystals for multiplexed biological labelings is marred by lack of general host/activator systems for efficient energy transfer for different dopant ions and lack of compelling approaches for demonstrated tunable emission with a broad range of color output by single wavelength excitation. Here, we present a general and versatile approach to fine-tuning the down- and up-conversion emission colors of lanthanide-doped nanocrystals, based upon the selection of appropriate activator/host systems. By precise control of emission wavelengths and intensity ratios through choice of different

combinations of dopant ions (Eu^{3+} , Sm^{3+} , Dy^{3+} , and P^{5+}) and dopant concentrations, downconversion emission of the YVO_4 nanocrystals can be easily tuned under single-wavelength excitation using a hand-held UV lamp. In a similar manner, upconversion multicolor modulation are realized in a single source of NaYF_4 nanocrystals doped with Yb^{3+} , Er^{3+} , and Tm^{3+} .

A00740-01311

Building an Artificial Cell Wall: Functional Biomolecule Assemblies in a Biomimetic Membrane Scaffold

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Harvesting the potential in the high selectivity and specificity of enzymes and transfer proteins for biosensing, biocatalysis and ion-selective filtration applications has long been in the forefront of nanobiotechnological research. As most of the biomolecules do not retain activity when removed from their native environment, mimicking physiological conditions is necessary. Remarkably, a large percentage of these proteins are associated to the cell membrane. Thus, establishing an artificial cell membrane scaffold might be sufficient to maintain enzymatic or transfer activity. Based on our previous works on the formation of biomimetic membranes and the design of an artificial cell, here we present the use of the biomimetic cell membrane scaffold to cover $\sim 1 \text{ cm}^2$ surfaces while incorporating functional protein assemblies. Membranes were deposited onto either a polyelectrolyte scaffold, as in the hollow sphere artificial cell, or onto a carboxylic acid terminated thiol SAM. Quartz Crystal Microbalance (QCM) measurements confirmed single bilayer coverage in both case. Gramicidin ionchannels were active in the scaffold. A biosensor/bioreactor candidate three member P450 redox enzyme assembly (c17, CPR and b5) was the incorporated into the membrane post deposition. Radioimmunoassay proved that the reconstituted complex exhibit biocatalytic activity; turnover by P450c17 was dependent on the amount of components of the b5 and CPR. These results suggest that the biomimetic membrane artificial cell wall supports the native function of membrane-associated biomolecules, and might be used for biocatalytic and, if electrochemical controll of the enzyme activity is implemented, for biosensor applications.

A00743-01316

Synthesis of II-VI Semiconductor Quantum Dots Applications for Live Cell Imaging

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We present the synthesis of II-VI semiconductors core-shell quantum dots (QDs) of CdSe/ZnSe and CdSe/ZnS and the surface modifications of QDs for live cell imaging. Cadmium oxide (CdO), diethyl zinc [$(\text{C}_2\text{H}_5)_2\text{Zn}$] and [$(\text{THS})_2\text{S}$] were used as sources of cadmium, Se and S, respectively. The core-shell QDs were found to exhibit enhancement in the luminescent efficiency compared to the core QDs as well as higher quantum yield (QY) and narrower full width at half maximum (FWHM). The particle size of core-shell QDs was observed to be $\sim 5 \text{ nm}$ by particle size analyzer (PSA) and transmission electron microscope (TEM). To prepare QDs for biological applications, the surfaces of QDs were decorated with various functional groups, such as mercaptopropionic acid (MPA), cysteamine (cys), dihydrolipoic acid (DHLA) and poly (ethylene glycol) (PEG) derivatives (like $-\text{OH}$, $-\text{NH}_2$ terminating with dithiol on other side), to make it water soluble. The surface modified QDs were used for living cell imaging in different cell lines such as HeLa and NIH/3T3 cells. It was found that the location of QDs in cells depended on their surface modifications.

A00750-01432

Surface Modification of Magnetite Nanoparticles and Their Intracellular Uptake

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Superparamagnetic iron oxide nanoparticles with suitable biocompatible molecules have been widely used for *in vivo* applications such as magnetic resonance imaging contrast agent, tissue engineering, immunoassay, detoxification of biological fluids, hyperthermia, drug delivery and cell separation etc. Surface-functionalized magnetite nanoparticles are suitable for receptor targeting because of their colloidal stability, biocompatibility, biodegradability, and non-toxicity for the body. In the present study, Iron oxide nanoparticles were prepared by the thermal decomposition of iron(III) acetylacetonate [$\text{Fe}(\text{acac})_3$]. Oleic acid (OA) and oleylamine (OAm) were used as surfactants to control the size of iron oxide nanoparticles. sulfo-SMCC were then coated onto the surface of the magnetite nanoparticles in order to introduce maleimide group on the surface.

Finally, homing peptide AP-1, which obtained by phage display technique, was immobilized on the surface of the sulfo-SMCC coated for specific targeting. The magnetite nanoparticles and peptide modified nanoparticles were then characterized by X-ray diffractometer (XRD), fourier-transform infrared spectrophotometer (FT-IR), transmission electron microscopy (TEM), dynamic light scattering (DLS) and vibrating sample magnetometer (VSM). Interaction of homing peptide-conjugated nanoparticles with bladder cancer cells were investigated to examine intracellular uptake of the particles into target cells.

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A00758-01337

Development of Biosensor for Detection of Mutations in DNA Using the Principle of FRET

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A biosensor for medical diagnostics using DNA sequence and mutation detection has been developed. The basic principle of this biosensor is that when a PNA probe labeled with a fluorescent dye (PNAC*) is hybridized with ssDNA having a sequence complementary to that of PNA, the hybridized complex acquires a negative charge and is electrostatically attracted towards the cationic conjugated polymer (CCP) present in the solution in water. If the concentrations of CCP (donor) and PNAC* (acceptor) are appropriate, the distance between the two may be within the critical radius required for transfer of electrostatic energy from donor to acceptor due to Forster resonance energy transfer (FRET). Under the condition of efficient FRET, the PL emission from CCP will be almost completely quenched and that from PNAC* will enhance significantly. If the ssDNA has sequence non-complementary to that of PNA, FRET will not occur or will be very weak. In the presence of mutations, the FRET signal is weak as compared to the case of complementary sequence. In the present work, we have tested our biosensor for Bacillus Anthracis for a 20mer sequence using polyfluorene phenylene (PFP) here. We have successfully detected DNA of Bacillus Anthracis for different number of mutations (15,8,5,4 bp) randomly oriented. We have also observed that the sensitivity of the sensor is improved by using a standard solution i.e. 1 x TE buffer (Tris HCl + EDTA). The results will be presented for the detection of 1-4bp mutated sequences with improved sensitivity. It is expected that the sensitivity of the sensor will be further improved by adding a non-ionic surfactant like n-dodecylpentaerythylene glycol ether (C₁₂E₅).

A00775-01361

The New Applications of Beta-lactam Antibiotics Derivatives in Imaging of Gene Expression and Detection of Drug Resistant Bacteria

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Since the antimicrobial properties of penicillin were first discovered in the last century, β -lactam based antibiotics have been extensively developed as miracle drugs in the treatment of diseases caused by bacterial infections in clinics. However, increased bacterial resistance to β -lactam antibiotics has been broadly documented and has now become a serious public health threat. The most common mechanism for β -lactam antibiotic resistance is the production of a family of bacterial enzyme: β -lactamases (Bla), a type of enzymes secreted both by gram-positive and gram-negative bacteria, which can efficiently cleave penicillin and cephalosporin based antibiotics before they can react with the target proteins in bacteria. How to rapid and sensitive detection of Bla in biological samples and highly efficient screening the drugs to inhibit the bacterial resistance before conducting effective therapy is thus of large importance in clinic. In terms of the important properties of Bla, the relevant studies have been exploited as following:

1) As a bacterial enzyme, Blas exhibit their novel properties such as their relative small sizes and monomeric structural feature which can easily express in eukaryotes cells without significant toxicity and endogenous interference. Such unique properties allow Bla to be used a reporter for examining the promoter/regulatory elements activity in cell biology. We developed a series of novel β -lactam based fluorescent probes for real-time imaging of β -lactamase expression in vitro, in single living cell and in the living animals.

2) In combination of unique gold nanoparticles (Au-NPs), we also present a novel and simple approach to efficiently identify β -lactamase activity and screen the enzyme inhibitors in a high throughput manner. This approach offers an alternative platform to rapidly evaluate the enzymatic activities and efficiently screen the inhibitors of β -lactamases in real time. It may also provide the practical application for rapid and specific detection of antibiotic resistant bacteria in clinical settings.

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A00785-01378

Development of Multi-Functional Electrochemical Oligonucleotide-based Sensors

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Electrochemical oligonucleotide-based sensors are of interest as they are inherently simple, possess rapid response times and can be designed not only detect its complementary strand but also other targets such as protein. Herein, we present two such sensors: (i) firstly for the detection of the complementary target at an alumina membrane by monitoring the blocking effect of a redox probe; (ii) secondly for the detection of protein thrombin at a gold substrate by tracking the electrochemically labeled, thrombin-binding oligonucleotides.

In the first sensor, a sensitive alumina membrane-based electrochemical oligonucleotide was fabricated. The alumina surface was covalently functionalized with probe oligonucleotide and the sensing mechanism is based on the binding the probe oligonucleotide and complementary target oligonucleotide, which blocks the approach of ferrocenemethanol towards the exposed platinum surface beneath the alumina layer. This results in incremental decrease in observed current with increasing probe oligonucleotide concentration. The sensor was applied for sensing of complementary oligonucleotide detection and found to have a low detection limit of 3.0×10^{-18} M target oligonucleotide as well as a linear dynamic range of 1×10^{-18} to 1×10^{-12} M.

Secondly, electrochemical monitoring of nanomolar concentrations of thrombin was performed using heterogeneous “switch-on” and homogeneous “switch-off” approaches that incorporated ferrocenyl aptamers assembled on gold substrates. For the first time, the heterogeneous approach was coupled to a glucose/glucose oxidase (GOx) amplification-regeneration system which increased its sensitivity by 2 folds with detection limits of 4.3 nM and 2.5 nM in the absence and presence of glucose/GOx, respectively. A new homogeneous system was also investigated which involved the ferrocenyl aptamer binding thrombin in solution causing a significant decrease in its diffusion coefficient. Thus the ferrocene anodic current decreased at an unmodified gold electrode with detection limit of 3.9 nM and 12 times larger linear range than the heterogeneous method.

A00807-01413

Synthesis of Silver Nanoparticles Using Acacia Gum as a Reducing Agent

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Synthesis of noble metal nanoparticles for applications such as catalysis, electronics, optics, environmental, and biotechnology is an area of constant interest. The design of a synthesis method in which the size, morphology, stability and properties are controlled has become a major field of interest. The conventionally available methods may consume a lot of energy in order to maintain the high pressures and temperatures that are needed for them to work. Various chemical and physical synthesis methods, aimed at controlling the physical properties of the particles, are currently employed in the production of metal nanoparticles. Most of these methods are still in the development stage and problems are often experienced with stability of the nanoparticles preparations, control of the crystal growth and aggregation of the particles. Most frequently, polymers are being used in the chemical synthesis of nanoparticles renowned as steric stabilizers or capping agents. In the early days of colloid science, plant extracts and gums were used routinely for stabilizing colloidal metal dispersions. This natural polymer not only is very low cost and abundantly available in nature but also has excellent emulsifying and surface-active properties, which can be beneficial for the design of metal nanoparticles.

In our present work we report a novel and convenient method for the rapid synthesis of silver nanoparticles using polysaccharide, acacia gum as a reducing and stabilizing agent under optimized conditions. The characteristic brown colored solution provided a convenient spectroscopic signature to indicate the formation of silver nanoparticles. UV-Vis absorption measurements in the range 350-600nm gave a deeper insight into the optical properties of the formed nanosized Ag particles by providing information about their size, size distribution, and surface properties. In the earlier study reported by Mohan et al.(2007) the characteristic surface plasmon (SP) resonance band of Ag nanoparticles centered at about 420 nm was observed after 24 h whereas in the current study, the band appeared after 5 min and attained the maximum intensity within 1 hour. By altering the reaction conditions, both the time and size of the nanoparticles could be tailored. The progressive enhancement of the SP band of Ag nanoparticles monitored over a broad spectrum indicated the increased yield and the high absorbance features. The Ag solutions were found stable even after 6 months as monitored by the absorbance. The coordination behaviors between amino groups in acacia gum and Ag^+ ions is responsible for the stabilization of the

silver nanoparticles. It can be concluded that the proteins or peptides form a coating over the silver nanoparticles, which in turn supports their stabilization.

The average size, size distribution, morphology, and structure of particles were determined by XRD, AFM, and UV/Visible absorption spectrophotometry. Fourier transform infrared (FTIR) study disclosed that the amino groups were bound to the particles accounting for the stability of the nanoparticles. The formed silver nanoparticles have a potential to be used in bioimaging and in biosensing.

A00842-01588

Studies on Technologically Important Carbonate-free Ferroelectric Nanostructures and their Biosensing Applications

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Ferroelectric materials are considered as potential candidates in ceramic and electronic industries. A ferroelectric material displays spontaneous polarization even in the absence of an external electric field below a critical temperature. Out of many ferroelectric materials, barium titanate (BaTiO_3) and lead titanate (PbTiO_3) are important systems in the sense that they are ferroelectric even above room temperature and display polarization values several orders more than the conventional potassium dihydrogen phosphate (KDP) type ferroelectrics. In this work, we highlight production of high quality BaTiO_3 and PbTiO_3 nanoparticles synthesized via solid state cum solvothermal approach. The solid state route is inexpensive and attractive for large scale production. We characterize the ferroelectric nanostructures by x-ray diffractometer (XRD), high resolution electron microscopy and differential scanning calorimetry (DSC) etc. In addition, high frequency response of polymer embedded ferroelectric nanostructures is discussed. Finally, we show that the ferroelectric nanocrystallites have antibacterial character and they can find useful application in life sciences and nanobiotechnology.

A00843-01466

Gold Nanoparticles with Tailored Size and Shape for Biosensing

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Size and shape control are hot topics in both Colloid Science and Nanotechnology. In the case of metals, size and shape control can be used as a means to tailor the optical properties through modification of the plasmon resonance condition. Many synthetic protocols have been published for metal nanoparticle synthesis, though simultaneous size and shape control are still rare.

We present here the controlled synthesis of extremely regular gold nanocrystals through ultrasound-induced reduction of HAuCl_4 on pre-synthesized seeds, using PVP as a stabilizing polymer. A strict relationship between the final morphology and the crystalline structure of the seeds has been observed, with formation of decahedra (pentagonal bipyramids) using penta-twinned Au seeds but single crystalline octahedra using single crystal Pt seeds. The dimensions can be strictly controlled through the ratio between the amount of seed and the HAuCl_4 concentration, and the monodispersity is as good as 10%. Additionally, the same procedure can be used to induce reshaping of gold nanorods, with the final morphology being again determined by the crystallographic structure of the initial rods.

The optical properties of these particles can be reproduced with a very good agreement by means of a model for the resolution of Maxwell's equations for bicones, so that a good correlation between particle size and optical response can be established, which will be extremely helpful to predict the optical response in biosensing applications. Examples of LSPR shifts induced by dielectric changes around various types of particles will be discussed.

A00853-04384**Biodetection and Diagnosis Strategies by Using SERS and SEFS**Ramon A. ALVAREZ-PUEBLA*Departamento de Química Física, Unidad Asociada CSIC-Universidad de Vigo, Vigo, Spain*

Surface-enhanced spectroscopies (SES) are a family of powerful analytical techniques that allows ultra-sensitive chemical or biochemical analysis and characterization. Both surface-enhanced Raman scattering (SERS) and surface-enhanced fluorescence spectroscopy (SEFS) require the generation of high electromagnetic fields at the surface of gold and silver nanostructures. Although SES have often been reported as ultrasensible techniques, several inherent problems due to the nanoparticle-analyte interaction or to the sample complexity may hinder the applicability of these spectroscopies to a wide range of relevant bio-problems. Herein we present several alternatives ranging from the direct detection, induced adsorption or indirect specific detection and labeling to overcome those drawbacks by selecting the appropriate materials and analytical media, and their use to the development of new platforms for high-throughput screening in real-time multiplexing devices.

A00854-01576**Lipid Encapsulation of Rare Earth-Doped Ceramic Nanophosphors for Fluorescence Bioimaging under Near Infrared Excitation**Kimikazu TOKUZEN¹; Kohei SOGA^{1,2};
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Fluorescence bioimaging is an important technique to visualize phenomena and migration of substances in biological systems by using fluorescent agents. Many of the problems in fluorescence bioimaging, such as damages on biological substances, color fading, autofluorescence and light scattering are caused by the irradiation of ultraviolet(UV) or visible(VIS) light for excitation. The use of rare-earth-doped ceramic nanophosphors as a fluorescent agent can solve the above problems because they are known to exhibit upconversion (visible) and efficient NIR fluorescence under NIR excitation. To apply them as biophotonic devices, the dispersion stability under physiological conditions, the control of specific interaction with a target substance and biocompatibility

must be introduced to the probe. Liposome is bilayer vesicle structured by phospholipids which have similar composition to biological membrane, and show low toxicity and antigenicity. Also, both hydrophilic and hydrophobic agents can be encapsulated in it and a variety of targeting ligands can be introduced on the surface of it. In this study, anionic liposomes encapsulating Er³⁺-doped Y₂O₃ nanoparticles were prepared from dipalmitoyl phosphatidylcholine (DPPC), dipalmitoyl phosphatidylglycerol sodium salt (DPPG) and cholesterol by using a complex emulsion method. NIR-to-visible upconversion emission was observed from the liposome under 980-nm NIR excitation in the field of biological microscope. According to zeta-potential measurement, the surface charge of the liposome encapsulating the Y₂O₃ nanoparticles is negative, while that of the Y₂O₃ nanoparticles is positive. FE-SEM observation revealed that the Y₂O₃ particles were encapsulated in thin polymeric capsules with approximately 650 nm diameter. We also introduced biotin and polyethylene glycol (PEG) to the liposomes encapsulating the particles by using DPPC, 1,2-distearoyl-sn-glycero-3-methoxypolyethylene glycol, 1,2-dipalmitoyl-sn-glycero-3-phosphoethanolamine-N-Biotinyl sodium salt and cholesterol in the above method. The specific interaction of the liposome to the avidin on a plate was evaluated. They could be dispersed in a physiological salt solution for more than 20 hours.

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A00876-02293**Controlled Synthesis and Biomedical Applications of Monodisperse Magnetic Nanocrystals**Yanglong HOU*Department of Advanced Materials and Nanotechnology
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Monodisperse magnetic nanocrystals have become tremendous interest due to their potential applications in magnetic resonance imaging (MRI), drug delivery, and bioseparation. Here we report the controlled synthesis of magnetic nanocrystals such as nanoparticles, nanocubes as well as multifunctional nanostructures. The biomedical applications of magnetic nanocrystals are also addressed.

A00889-01547

Core-shell Upconversion Fluorescent Nanoparticles for Imaging and Photodynamic Therapy of Cancer

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Use of NIR-to-visible upconversion fluorescent nanoparticles for biomedical applications introduces many advantages, for example, minimum photo-damage to living cells, weak auto-fluorescence, high detection sensitivity and high light penetration depth, etc. To date, lanthanide doped hexagonal phase NaYF₄ nanocrystals have demonstrated to be the best NIR-to-visible upconversion materials and hexagonal-phase nanocrystals have higher upconversion efficiency than cubic phase nanocrystals. However, synthesis and use of this material is still in its infancy due to poor water dispersibility and undesirable surface property. It is important to develop efficient and convenient methods for synthesizing high-quality and monodispersed NaYF₄ nanocrystals with functionalized surface for various biomedical applications. In our lab, an efficient and user-friendly method has been developed for synthesis of hexagonal-phase NaYF₄ nanocrystals with very uniform silica coatings on the surface. NaYF₄ nanoplates, nanospheres and nanoellipses are produced and all these nanocrystals show strong upconversion fluorescence. The silica coated nanoparticles are well dispersed in water to form a transparent colloidal solution and the surface of the nanoparticles can be functionalized for biomolecule conjugation. The nanoparticles have been used in bioimaging as fluorescent probes.

Using fluorescent imaging for cancer diagnostics has resulted to a new type of therapy known as photodynamic therapy (PDT). Two non-toxic components are required in PDT: photosensitizer, a photosensitive molecule that localizes to a target cell and/or tissue, and administration of light of a specific wavelength that activates the sensitizer. The photosensitizer transfers energy from light to molecular oxygen, to generate reactive oxygen species which can kill cancer cells. PDT has been proved to be effective in early lung cancer, bladder cancer, head and neck cancers, and to be the ideal treatment for skin cancer, however, the visible light needed to activate most photosensitizers usually can not pass through a thick layer of tissue. For this reason, PDT is usually used to treat tumors on or just under the skin or on the lining of internal organs or cavities, and is less effective in treating large tumors. In this work, a photosensitizer, zinc phthalocyanine (ZnPC), is incorporated into the porous silica coated onto NaYF₄ upconversion nanoparticles, and the effectiveness of the photodynamic therapy is assessed. The upconversion

nanoparticles can convert NIR light to visible light which then activates the photosensitizer.

A00907-01595

Fluorescence and Two-photon Absorption Properties of Organic Nanoparticles

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Nanoparticles are very attractive materials. Nanoparticles of metals and semiconductors have properties different from crystals or atoms. Energy of an electron in a metal or semiconductor nanoparticle depends on the particle size significantly due to quantum effects. On the other hand, properties of organic nanoparticles have not been fully clarified yet. In these twenty years, optical properties of fine particles mainly particles with micrometer size, of organic compounds have been studied. Recently, a new method to produce nanoparticles has been developed. This method has many advantages compared with other methods. One of them is materials insoluble to solvents can be applied. In this study, we measured visible absorption spectra, fluorescence spectra, fluorescence quantum yields, and fluorescence lifetimes, of organic nanoparticles. Fluorescent organic nanoparticles may be used for bioimaging. We also measured two-photon absorption cross sections of organic nanoparticles.

Naphthacene, anthracene, 9-methylanthracene, 2-ethylanthracene, perylene, Alq3 [tris-(8-quinolinolato) aluminum], bis-MSB [1,4-bis(2-methylstyryl)benzene], Zn(Mq)2 [bis(2-methyl-8-hydroxyquinolinato)zinc], BBO [2,5-di-(4-biphenyl)-oxazole], dimethyl-POPOP [1,4-bis(4-methyl-5-phenyl-2-oxazolyl)benzene] were used to prepare nanoparticles for fluorescence measurements. Transparent organic nanoparticles dispersed in water were prepared by reprecipitation method and a method of laser ablation in water. Although almost all organic compounds we studied did not fluoresce in nanoparticles probably due to concentration quenching, fluorescence quantum yield of anthracene nanoparticles was as large as 0.46. Nanoparticles of Alq3 shows weak fluorescence. Naphthacene or perylene doped anthracene nanoparticles show relatively high fluorescence quantum yields. Fluorescence lifetimes of organic nanoparticles are very short or shorter than those of dye molecules in THF solution.

We also measured two-photon absorption cross sections of nanoparticles of organic dyes, such as perylene derivatives, by the open aperture Z-scan method using a femtosecond Ti: sapphire regenerative amplifier laser. Transparent organic nanoparticles dispersed in water were prepared by a method of laser ablation in water. A surfactant, sodium

dodecyl sulfate, was added to stabilize the colloidal solutions. Two-photon absorption spectra were measured from 780 to 840 nm. Methanol solution of rhodamine 6G was used as a reference of two-photon absorption cross sections. The values of two-photon absorption cross section per molecule of nanoparticles were 34 GM (1 GM = 1×10^{-50} cm⁴ s/photon) for PTCDA, 94 GM for PTCDI, 129 GM for perylene-66, and 19 GM for quinacridone. All studied compounds have a maximum of two-photon absorption cross section at 820 nm in the measured wavelength range of 780-840 nm. Nanoparticles of perylene-66 have a little higher two-photon absorption cross sections than the molecules in tetrahydrofuran. Although nanoparticles with high fluorescence quantum yields and large two-photon absorption cross sections will be very useful for bioimaging, we have not found good nanoparticles yet. We are now studying nanoparticles made of two compounds.

A00920-01617**Dendron-Modified AFM Probes for Sensing DNA and Proteins and Mapping mRNA on Tissue Surface**

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A second-generation dendron (or 9-acid) and a third-generation dendron (or 27-acid) have been applied to AFM probes to control spacing between the immobilized biomolecules on them. For DNA-DNA interaction, both binding and unbinding events were observed, and enhanced sharpness of the force histograms was an additional advantage from the controlled surface. With the same approach, the interaction between the signal transducing proteins were examined, and quantitative information at the single molecule level was obtained. Also, distribution of pax6 mRNA in mouse embryonic neocortex has been mapped with the DNA-modified tips. In order to understand the interaction between RNA and DNA, two different model systems were studied, and the investigation on the tissue surface showed that mapping was in harmony with the fluorescence mapping. Especially the new approach detected mRNAs in the region that the conventional analytical tool could not see.

A00976-01695**Synthesis and Characterization of Gold Nanoparticles via Reduction Methods with Different Reducing Agent**

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Various concentration of Na salt L-glutamic acid was used to prepare gold nanoparticles by the reduction of aqueous gold chloride with water. The as-synthesized gold nanoparticles were characterized by ultra violet-visible absorption spectroscopy (UV-Vis) and transmission electron microscopy (TEM). Samples with 25mM and 35mM of amino acid derivatives show absorption peaks at 519 nm, while sample with lower concentration, 15mM and 5mM show absorption peaks at 520nm. Gold nanoparticles prepared by the Turkevitch method shows the absorption peak at 534nm. The TEM analysis for gold nanoparticles with amino acid derivatives shows a size of gold nanoparticles in a range of 13 nm to 16nm. Comparison of amino acid protected gold nanoparticles capped with and without chitosan is also performed.

A01009-04609**Applications of Carbon Nanotube Networks in Biomolecular Detection**

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Most DNA detection methods rely on optical, piezoelectric, or electrochemical transductions. The detection based on the resistance change of a single silicon nanowire or an individual carbon nanotube has also been reported. However, these sensors may have significant device-to-device variations. Recently, field-effect transistors (FETs) based on single-walled carbon nanotube (SWNT) networks have been fabricated and their electrical properties depend on the percolation paths of SWNTs in conduction channels, where device variations are expected to be small. These devices are potential for label-free detection of biomolecules. Herein, we take detection of DNA hybridization as an example to discuss the detection mechanisms (metal-SWNT junction vs. channel) for these types of devices. The detection limit can be enhanced from 1nM to ~100pM by using a "nanoparticle-reporter enhancement" approach. Another advantage of SWNT network based devices is that the devices can be easily extended to micron or even mm long in channel length. Therefore it is particularly useful for monitoring the activities of living cells. Examples will be given to demonstrate the dynamic detection of biomolecular secretion from living cells.

A01166-01993

A Simple Method Towards Self-Assembled Superparamagnetic Quantum Dots

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Given the disastrous consequences of neurotrauma, and controversy surrounding current treatments, we aim to deliver drugs to neurotrauma injury sites using nanoparticles. We have prepared a composite nanomaterial incorporating cadmium telluride (CdTe) and magnetite (Fe_3O_4) nanoparticles. The luminescent properties of CdTe will be used to enable tracking *in vitro* and *in vivo*, while magnetic characteristics will facilitate spatial positioning of the delivery agent. The aqueous synthesis of CdTe quantum dots was achieved using microwave energy, while Fe_3O_4 was prepared by aqueous co-precipitation. The combination of these materials to form the nanocomposite was performed in a simple manner via self-assembly. We find that this material combines the benefits of both its components: sharp emission spectra and high saturation magnetisation, and that the superparamagnetic properties of Fe_3O_4 are maintained in the composite. We will present details on the preparation and properties of this material.

A01178-02041

Nanoporous Electromembrane in Bioanalysis

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Recent advances in the synthesis of nanomaterials and chemical functionalization of these nanomaterials, have accelerated the developments of bioanalysis. As we progress towards molecular level analysis, the physico-chemical nature of elements at the interface functioning as signal transducers in the case of sensors or facilitating selective separation in the case of separation techniques will become increasingly crucial to overall performance of analytical techniques. Design and fabrication of advanced materials coupled with good understanding of their behaviors when incorporated as interfacial or transducer elements is necessary to the process of bioanalysis using sensor or separation techniques. In this presentation, we will look at some of the recent developments of biosensors using nanoporous membrane coated electrodes.

Second, we will additionally present the work on electrode-membrane-electrode system first developed by our group which has been demonstrated for use in controlling transport of nanoparticles, proteins and electroactive species. The nanoporous membrane was made conductive by physical sputtering of metal on both sides of the membrane. Electrical potential difference is applied across

the membrane through these metal layers which could also function as sensing electrodes. Advantages include rapid detection of electroactive species at the membrane surface and direct correlation of signals to concentration of analytes at the membrane surface.

A01197-02145

Nanostructures for Bio-Chemical Sensors

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Nanotechnology is bringing unprecedented opportunities to biology. The nano-structured materials such as nanoparticles, nano-pores, carbon nanotubes and nanowires, arranged in numerous configurations are playing an increasingly important role in the development of bio-chemical sensors. Different well established process techniques like chemical vapour depositions in the presence of catalysts or standard silicon processes like lithography, etching etc are being exploited for the creation of these nano-structured devices. The nanoscale features allow the intimate interactions of the analytes within devices, which can improve the sensitivity and performance of the sensors. The use of these nano-materials has allowed the introduction of many new signal transduction technologies in biosensors.

This paper presents the over-review of various nanostructure-based bio-chemical sensors with particular emphases on the one-dimensional silicon nanowires configured as field-effect transistor (FET) and ordered nano-spaced silicon structures for Surface Enhanced Raman Spectroscopy (SERS).

A01199-02048

Detection of Thiols by MOS Capacitor for Biosensor Applications

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Thiols attached to gold surface provide the platform for many biosensors to detect DNA. Attachment of thiol to gold can create stress in the gold, especially if the layer is thin, resulting in a change in its work function. Kelvin probe technique has been used earlier to measure this change in the work function of gold. Here, we report a new and simple method of using a gold gated MOS capacitor to monitor the change in the work function of gold due to thiol attachment as a flat band voltage shift in the capacitance-voltage (CV) characteristics. MOS capacitors are relatively simple to fabricate as compared

to a MOSFET or MEMS cantilever, and can therefore be used as effective biosensors.

MOS capacitors of area $450\ \mu\text{m} \times 450\ \mu\text{m}$ with 1-10 $\Omega\text{-cm}$ p type (100) silicon, 40 to 50 nm silicon dioxide and 30 to 50 nm thick gold were fabricated. The capacitance voltage (CV) measurements were done using the Agilent E4980A LCR meter at 100 kHz and 1 MHz with an ac bias of 50 mV, sweeping the dc bias from -5V to 2V.

The samples were kept immersed in 1 - 10 mM solution of 1-octadecanethiol (ODT) in ethanol for varying duration of up to 24 hours followed by rinsing in ethanol and drying with nitrogen gas. The presence of thiol on the surface was verified by monitoring the contact angle which confirmed the hydrophobic nature of gold surfaces modified by thiol.

To study the effect of the duration of immersion, flat-band voltage and contact angle were measured on samples immersed for different durations in 1-octadecanethiol and it was observed that the monolayer coverage on the surface of gold increased with time. The effect of concentration of thiol solution on the shift in flat-band voltage was also studied and it was observed that there is saturation in the flat-band shift with concentration of the solution.

The experiments were repeated for 4-fluorothiophenol and a shift in CV curves towards the right was found.

We have demonstrated that a gold gated MOS capacitor can be used to sense the attachment of thiol molecules to the gold. The results from the CV measurements were corroborated with measurements of contact angle. This simple technique can be extended to sense attachment of DNA to the thiol and is being currently investigated.

A01273-03413

Single Walled Carbon Nanotubes (SWCNT) for Herbicide Detection

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Single-walled carbon nanotube (SWCNT) is a one-dimensional system with all its carbon atoms present on the surface, hence its conductance is highly sensitive to the surrounding charge environment. Due to the extreme charge sensitivity, biocompatibility and chemical stability,

SWCNT is particularly interested in sensing application. One of the increasing demands in signal detection is required in agricultural industry, where the usage of herbicides has increased tremendously to improve agricultural production in order to meet the world population demand. While the introduction of herbicides has successfully prevented crop losses before and after harvesting, it poses a serious threat to human health. The residues from herbicides can remain in soils for certain period of time and readily be absorbed into food and water source, which can become more concentrated as they moved up the food chain.

In the present work, the potential application of carbon nanotubes in herbicide detection was investigated based on direct electrical detection. Atrazine (2-chloro-4-ethylamino-6-isopropylamino-1,3,5-s-triazine), a member of s-triazine herbicides which is widely used to control broad leaf weeds in corn, sorghum, sugarcane and etc, was tested using liquid gated carbon nanotube transistor. All measurements were carried out in a liquid environment to allow real time detection.

While AFM images showed strong interaction between the SWCNT network and the molecules, the electrical detection of unmodified atrazine to its corresponding antibody gave insignificant changes. On the other hand, by conjugating the atrazine molecules with carrier protein, bovine serum albumin (BSA), a decent change in the current level was observed. This observation, combines with our result of unmodified atrazine, give a strong indication that the working mechanism behind the liquid gated CNT transistor is determined by the changes in the electrostatic environment surrounding the SWCNT.

A01302-02562

Photosystem II-Based Biomaterials for the Detection of Pollutants

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During several decades, uses of herbicides and organic compounds in agriculture are continuously increasing. The industrial effluents and heavy metals are making the condition worst. We need devices to detect these pollutants. Several researches have shown the development of various types of sensors but an efficient, cost-effective and portable sensor is still required for the easy monitoring of water pollutants. We are attempting to screen out water pollutants. We employed Photosystem II particles as a biosensing material which is the main target of most of the herbicides, some heavy metals and organic pollutants. They are biologically active materials

which are capable of oxygen evolution by hydrolysis of water in the presence of light. This property is most useful because its physiological activity can be easily monitored by amperometric/potentiometric systems. A single celled cyanobacterial species is like chloroplast of higher plants. We have attempted using whole cells and permeabilised spheroplasts as a biosensing material for biosensor designing. We have observed the changes in their spectral properties, peak shiftings, electron flow rate and oxygen evolution properties. The output results were available in a short time span. The biomaterials were immobilized on the working electrodes and Silicon wafers and can also be immobilized on any kind of flow cell system transducers.

A01308-04290

Non-covalent Interaction of Single Walled Carbon Nanotubes with Ethylene-vinyl Acetate Copolymer Conductive Composite Films for Sensor Application

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Chemically functionalized single walled carbon nanotubes (SWNTs) were useful in material science because of their potential ability to bind to organic and polymer matrices either through van der Waals interactions, hydrogen bonding, or covalent bonding. In this endeavor, a judicious selection of a polymer and processing methodologies are important for uniform dispersion of SWNT into polymer matrix. A number of non-covalent modified SWNT dispersed ethylene-vinyl acetate (EVA) copolymer and maleic anhydride-grafted-EVA composites films were prepared with different composition of SWNT (0.1, 0.2, 0.5, and 1.0 wt %). These composite films have shown significantly improved properties such as high tensile strength, modulus, heat distortion temperature, barrier properties, UV resistance, and conductivity. The dispersed SWNT/polymer composite films were characterized by atomic force microscopy (AFM). The uniform dispersion of SWNT's in polymer within the percolation threshold limit and prominent increase in the electrical characteristics by I/V method. These SWNT/polymer composite films may also useful in magnetic and sensor applications.

A01358-02385

Self-assembling of Novel Multifunctionalizing Oligoarylenedithiols on Plasmonic Metal Nanoparticles for Label Free Bio-sensing

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Biorganic functionalization of gold thin films and nano cluster mesoscalar assemblies and their resulting optical properties have immense applications ranging from biosensing to nano medicine. The appealing properties are in the surface plasmon resonance of those bio-metal ensembles that can be tailored not only by the gold nanoclusters geometry but also by the nature of the ligating molecules and by the chemistry of the interface. This characteristic is being exploited for realizing nanoscale optical biosensors based on localized surface plasmon resonance of metal nanoparticles.

In spite of the fact that a significant level of understanding of the formation, aggregation and properties of self-assembled-monolayers (SAMs) has been gained, the detailed mechanism of their formation and how this depends on the structure of the functionalizing molecule is still not fully understood. Specifically, using thiols SAMs, the charge transferred during the SAM formation dictating its geometry and the fate of hydrogen atoms are unclear.

In this contribution we discuss the surface chemistry and kinetics and dynamics of interface phenomena playing a role during functionalization of plasmonic gold nanoparticles supported on Si(100) substrates and of semiconductor nanostructures such as Si and ZnO nanowires and nanorods.

A novel class of oligoarylenes bearing two thiol groups on one end of the conjugated chain and various functional groups on the other end has been synthesized. The relative position of the two SH groups offers an unusual binding geometry of the molecules to the gold surface, and the substituents attached to the other end of the conjugated chain enable further functionalization, including introduction of bio-molecules.”

The surface plasmon resonance of the metal nanoparticles is enhanced when they are coupled to the semiconductor nanowires and nanorods.

Those systems are used for further anchoring of bio-molecules, e.g., albumin and antibodies for biosensing and anchoring of porphyrins for gas sensing to NO.

Plasmonic spectroscopic ellipsometry (HORIBA UVISEL, Jobin Yvon) is exploited for the optical monitoring of the functionalization of metal nanoparticles and anchoring of the biomolecules to the semiconductor nanostructures. For such plasmonic metal enhanced semiconductor nanostructures-based sensors, the degree of coverage by the porphyrin and/or protein, the aggregation, the type of binding and orientation of bio-molecules on the semiconductor nanostructures surface is important for the sensing activity. Therefore, plasmonic ellipsometry is used to investigate the interplay among the nanostructures size and shape, the functionalizing solution concentration and dipping time, the mechanism of anchoring of the functionalizing molecules.

Monitoring of the surface plasmon resonance of the metal nanostructure coupled to the semiconductor is also used for label free sensing of the NO gas as well as of the gene-antigene interaction.

A01377-02402

Fabrication of Arrays of Sub-50-nm Nanofin structure via Photolithography and Nanocoating

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Fabrication of nanopatterns and nanomaterials with the dimensions of sub-50 nm has been actively studied, because such nanopattern and nanomaterials show exotic properties. Although photolithography-based fabrication is the most process-friendly approach, it requires very high cost for processing equipments and large space. This approach is now getting close to technical and process limitation, and alternative methods are, thus, strongly desired for sub-50-nm fabrication.

We herein introduce new fabrication process to create nanostructures based on the combination of photolithography and the nanocoating. Especially, we focus on the preparation of nanofin shape on a substrate, since well-designed nanofin structure gives wide surface area which will be effective for sensing target analytes. In our process, a photolithographically fabricated line template on a silicon wafer was coated with a nanolayer, and then the topmost portion of the coating layer and the template were successively removed using plasma gases, leaving the sidewalls of the coating layers remaining on the substrate. Even the thickness of the fin is less than 50 nm, these fins were fully self-supporting. The thickness of fin can be easily changed by adjusting the thickness of the coating layer. The height/width ratio of the fin is more than 20 which is surprisingly high when compared to that of the conventional photolithography process.

Based on this idea, we extended this process to prepare various nanoparts, aiming at the creation of functional nanomaterials. These results will be also discussed in this presentation.

A01397-02429

LSPR Based Biosensing on DNA Hybridization using Superstructured Au Nanoparticle Arrays Fabricated by Block Copolymer Self-Assembly

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The success of solid-supported array technology for high-sensitivity and high-throughput nucleic acid analysis has prompted many to envision analogous protein-based arrays for immunoassays and a range of protein expression investigations. In particular, Microarrays have conventionally been patterned by spotting, printing, or lithography techniques. To enable DNA and protein nanoarrays over large areas, the use of self-assembled templates, such as block copolymers (BCP's), may offer an attractive nanopatterning approach. In this presentation, we demonstrated that the BCP-based method enables two-dimensional high-density biosensor.

Noble metal, such as gold (Au) and silver (Ag), nanoparticles (NPs) exhibit unique optical properties due to the localized surface plasmon resonance (LSPR). Since the LSPR band is sensitively affected by the dielectric environment, biosensing based on LSPR has been actively utilized for the adsorption of infinitesimal amounts of biomolecules on the surface of metal NPs. Here, we present BCP-based strategies for patterning biosensing arrays on the nanoscale.

DNA nanoarrays based on plasmonic Au NPs arrays are created using self-assembled polystyrene-block-poly(4-vinyl pyridine) micellar films. The diblock copolymer micelles containing HAuCl₄, a precursor of Au NPs, are coated on a solid substrate to form a monolayer in a hexagonal order. Once the organic polymer is removed by oxygen plasma treatment, ordered patterns of Au NPs are fabricated. The overall procedure to generate such Au NP arrays was investigated by AFM study. The arrayed Au NPs are functionalized with a spontaneously self-assembled alkanethiol monolayer with a biotin moiety onto which streptavidin molecules are adsorbed for the immobilization of PNA probe. The biotinylated PNA probe is bound to the modified Au NPs surfaces via biotin-streptavidin interaction, and then the target material, complementary DNA molecule, is hybridized with the PNA probe. Such binding event was investigated by monitoring the changes of LSPR signal using both static and in situ methods.

Upon the adsorption of each layer, the LSPR signal was consistently changed in terms of both intensity and peak position. The results show that LSPR band responds to the stepwise buildup of each layer with high sensitivity.

The Au nanoarray-based sensor thus prepared is proved to be facile, simple, and reproducible to implement. Further, we also fabricated stacked, multilayers of Au nanoparticles arrays by repeating the deposition of Au-loaded BCP micelles, and compared its function as biosensory platform with monolayer Au arrays. Thus BCP self-assembly is demonstrated to be a versatile and straightforward strategy for arraying a variety of biomolecules on the nanoscale for solid-supported sensing technology.

A01415-02453

Oligonucleotide-based Sensing or Manipulating Elements

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Oligonucleotides are useful as a nano-material, especially as a sensing element. They worked not only as a genetic component but also as a ribozyme or DNAzyme instead of enzyme. Here we developed new oligonucleotide-based probes for detection of cellular RNA and prepared a new photo-responsive RNA aptamer by *in vitro* selection technique.

First, we designed a reduction-triggered fluorescence (RETF) probe that shows a high S/B ratio for sensing oligonucleotides. A new fluorescence molecule, rhodamine azide, that we synthesized is activated only by a specific reducing reagent on the oligonucleotide target and is very stable under biological conditions, showing little background fluorescence. The chemistry to activate the compound involves the reaction between the azide group of rhodamine derivatives and reducing reagents, with the fluorescence signal appearing after reduction of the azide group. The signal/background ratio of this fluorogenic compound reached 2100-fold enhancement in fluorescence intensity. Dithio-1,4-threitol or triphenylphosphine as reducing reagents were successfully utilized for this chemistry to be introduced into the DNA probe. The genetic detection requires that two strands of DNA bind onto target oligonucleotides, one probe carrying a reducible fluorogenic compound while the other carries the reducing reagents. The reaction proceeds automatically without any enzymes or reagents under biological conditions to produce a fluorescence signal within 10–20 min in the presence of target DNA or RNA. The probes were successfully applied to the detection of oligonucleotides at single nucleotide level in solution and endogenous RNA in bacterial cells.

Next, we designed new oligonucleotide-based aptamers that have been engineered through repeated rounds of *in vitro* selection or equivalently, SELEX to bind to various molecular targets such as small molecules, proteins, nucleic acids, and even cells, tissues and organs. Aptamers offer advantages over antibodies as they can be engineered completely in a test tube, and are readily produced by chemical process. By using this advantage, we have developed RNA and DNA aptamers containing natural and nonnatural components for targeting hemin. In particular, hemin-DNA aptamer complex as a biocatalyst, has been used for sensing, nanobiotechnology applications. Here, development of a photoresponsive RNA aptamer was attempted. We synthesized azobenzene-modified nucleotides, which can potentially be incorporated into a selection process. Subsequently, *in vitro* selection of photo-responsive RNA aptamer to hemin was carried out using the azobenzene-modified nucleotide instead of natural nucleotide. After 8 rounds of selection process, we obtained azobenzene-modified RNA aptamers which can bind hemin after irradiation of visible-light. One of the aptamers significantly enhanced hyperchromicity of the So-ret band of hemin by complex formation. Interestingly, after irradiation of UV light to the complex of RNA aptamer-hemin, the hyperchromicity of Soret band was significantly reduced. These results suggest that the RNA aptamer can associate or dissociate with hemin in response to visible or UV light, respectively.

A01430-03036

Tunable Light Emission Using Quantum Dot Coated Upconverters

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Fluorescent upconverting crystals (UCs) typically consist of metal oxides doped with lanthanide ions. When excited in the near infrared (NIR), the ions upconvert either through a non-linear 2-photon absorption process or by sequential absorption of photons via the formation of a long lived intermediate state. Such upconverters have a variety of potential uses in solar energy conversion and optical labelling,¹⁻³ however they are currently limited by the narrow range of emission wavelengths available. One solution to this problem is to use semiconducting nanocrystals or quantum dots (QDs) to tailor the UC emission characteristics.

Quantum dots are well known for their size tunable emission properties. In principle the combination of strong up-converter absorption with tunable down-conversion from semiconducting QDs provides a method for creating tunable upconverters. We demonstrate a simple method for encapsulating NIR upconverters such that they can emit at any tunable visible wavelength with a high emission quantum efficiency. By coating conventional NIR upconverters with multilayers of QDs, the upconverted emission is absorbed by the QD coating (i.e. radiative energy transfer) which then re-emits at its design (visible) wavelength, a feature that is not available with current lanthanide based upconverters.

The resulting tunable fluorophores are extremely photostable and can be transferred into glassy or polymer matrices. This process cannot generally be exploited with molecular species such as dyes because of their smaller absorption cross-sections. Importantly, the absorption cross-section of the QDs is high enough that just a few monolayers on the UC surface are sufficient to ensure both a high yield of radiative energy transfer and emission efficiency.

A01452-02520

Concerns for Biototoxicity of the Nanomaterials

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Nanotechnology has captured the imaginations of scientists, engineers and economists due to the numerous discoveries at the nanometer scale level and their potential applications and societal implications. This new technology, which uses nanomaterials is expanding rapidly and is affecting many aspects of everyday life. There are already hundreds of products that utilize nanomaterials and one can safely add that what one can do with this technology is almost like a science fiction. However, the rapid progress in development and uses of nanomaterials is not yet matched by toxicological investigations and there is a rapidly growing concern about the toxicity of these particles for the bio-environment surrounding the humans and the humans themselves. Several epidemiological studies have, in the past, implicated ultra fine particles polluting the air in the exacerbation of cardio-respiratory disease and increased morbidity.

In relation to the nanomaterials, studies conducted in experimental animals have shown that the unique chemistry and functionality of nanomaterials along with the increased concentration and higher reactive surface area per unit mass is important in the acute inflammatory

and chronic response. In addition, nanomaterials deposited in organ may enter the vasculature and hence reach other organs. The biological and systemic consequences of the exposure to higher levels of nanomaterials cannot be predicted since each material depend on its composition and reactivity. Hence there is a need to understand and clarify the toxicity or otherwise of each nanomaterial to enjoy the benefits of nanotechnology without being concerned about the exposed to the hazards of nanomaterials, which are unavoidable.

We have initiated studies using ZnO₂ nanoparticles and investigations on the absorption, transport and toxicity are in progress. This paper will discuss the concerns related to biotoxicity of nanomaterials, and what must be done to achieve maximum benefits of nanotechnology with least damage.

A01521-02654

Detection of E.coli Using an Electrochemical Immunosensor

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In this project, a membrane-based electrochemical immunosensor based on nanoporous alumina-modified platinum wire electrode was developed for the detection of Escherichia coli in environmental samples. The principle of this sensing mechanism is based on the blocking of the pore channels of the alumina substrate when the E.coli cells are specifically bound to the anti-E.coli physically adsorbed onto the channel walls, thus impeding the diffusion of the redox probe, ferrocenemethanol, to the platinum wire overlaid by the alumina substrate. The electrochemical immunosensor was evaluated for its quantitative response towards E.coli and a calibration plot relating the change in signal response towards varying E.coli concentrations is optimized through the study of variables involving antibody concentrations, pH, ionic strength and alumina pore sizes. Understanding of the sensing process is carried out using impedance studies.

A01540-02689

Positively Charged Silver Nanoparticles for Surface Enhanced Resonant Raman Spectroscopy of Dye-Labelled DNA

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Silver nanoparticles have been used extensively for detection of molecules by SERS (surface enhanced Raman spectroscopy). Raman spectroscopy has become an interesting option for multiplexed homogeneous assays, as the peaks of the Raman spectra are much sharper than fluorescence spectral peaks. Specifically, dye-labelled DNA molecules have been used for biosensing applications based on SERRS (surface-enhanced resonant Raman spectroscopy). However, because the DNA is negatively charged and the silver nanoparticles used for SERRS are usually negatively charged, a charge neutralization agent, like spermine, is added in order to adsorb the labelled DNA to the nanoparticles surface.

An interesting alternative, is to use positively charged silver nanoparticles. In such a case, the negatively charged DNA should theoretically act as the aggregating agent for the nanoparticles, without the need of additional compounds.

For this purpose we synthesized silver nanoparticles with a positive surfactant capping. High SERRS signals were generated in systems that included the positive particles and dye-labelled DNA, without the need of additional aggregating agents.

The addition of negatively charged nanoparticles was found to increase the kinetics of SERRS signal generation, and generate higher signals than those with positive particles/DNA only. The shape of the kinetic curve for SERRS signal generation was found to be qualitatively different from those seen in systems where negatively charged nanoparticles and charge-neutralizing agents are used.

In conclusion, positively charged silver nanoparticles can be used in a homogenous assay format for the detection of dye-labelled DNA without the use of charge-neutralization agents. They are promising new candidates for high-multiplexed DNA assay based on SERRS.

A01580-02754

Highly Luminescent Water-Soluble InP Nanocrystals with Thick ZnS Shell Prepared via Reactive Phase Transfer and Photochemical ProcessingNorio MURASE¹; Chunliang LI¹; Masanori ANDO¹; Hiroyuki ENOMOTO²*1. Photonics Department, National Institute of Advanced Industrial Science & Technology(AIST), Osaka, Japan**2. Graduate School of Engineering, Osaka Electro-Communication University, Osaka, Japan*

Semiconductor II-VI nanocrystals (NCs), such as CdSe and CdTe NCs, have been studied intensively in this decade. However, their toxicity prevents them from using commonly especially in bioimaging and biosensing fields. Water solubility is another important factor for these bioapplications.

In this talk, we will present a newly developed preparation method of highly luminescent water-soluble InP nanocrystals. The thick ZnS shell (more than 1 nm) is crucial for getting high emission efficiency (30-68%) in water.

InP NCs (2-4 nm) were prepared solvothermally in organic phase using a facile and safe method. These NCs were then transferred into alkaline aqueous solution containing thiol and zinc ions. When these NCs in aqueous solution were subsequently irradiated by ultraviolet (UV) light, they showed intense size-dependent photoluminescence (PL) from green to red due to the formation of a thick ZnS shell on the NCs. The surface dissolution of the NCs, under conditions in which bulk InP does not dissolve due to its covalent bond nature, was observed at two steps: phase transfer and shell formation. This dissolution competed with the formation of the ZnS layer at the start of UV irradiation.

Since the UV irradiation enables creation of a thick shell by optimizing the synthesizing conditions, the high PL efficiency was obtained in water. Quantum mechanical calculations explained this high efficiency as a result of confinement of electron in the NCs having a thick ZnS shell. The PL decay behavior of these water-soluble InP/ZnS NCs did not show obvious size-dependence, unlike HF-treated ones previously prepared in organic solution. This is attributed to the well-passivated surface states of the NCs due to their thick ZnS shell. The NCs showed a significantly higher In/P ratio than those previously reported. This indicates that In ions were preferentially located on the surface of the InP core in the NCs prepared by this method.

The NCs thus prepared in aqueous solution were stable for months in air and could be incorporated into a glass matrix using a sol-gel process with maintaining their spectral properties.

This study was supported in part by the Creation and Support Program for Start-ups from Universities, sponsored by the Japan Science and Technology Agency (JST).

A01596-02776

Fabrication of Biocompatible Nanoparticles for Molecular Imaging and Drug Delivery

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The talk describes recent advances in the synthesis of highly luminescent and magnet nanoparticles. It is shown that preparing the particles under continuous flow conditions allows reproducibility far beyond the limits of batch synthesis.

Special emphasis is put on the development of a protocol for ligand exchange against α,ω -substituted polyethylene-oxide containing ligands. It is shown that small changes in the composition result in tremendous differences in stability against serum and buffer solutions and fluorescence quantum efficiencies in the biological environment. We present types of ligands allowing reproducible and stable fluorescence properties of quantum dot systems.

We further present in-vitro and in-vivo data on cell toxicity and molecular imaging with such modified quantum dots and superparamagnetic iron oxide nanoparticles.

A01599-02781

Synthesis of Ternary ZnCdTe Quantum Dots with Highly Red Photoluminescence Properties

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There is an increasing interest in using quantum dots (QDs) in variety of applications such LED and solar cells due to their unique optoelectronics properties. Ternary QDs might be potentially used in currently existing application due to their novel properties in comparison to the binary system. Here, we report the synthesis of ternary QDs system by quick injection of chemical precursor into the reactor at moderate temperature (300°C). The ternary QDs have been synthesis using wet chemical process giving highly red emission with high photoluminescence after exposing under UV lamp (365 nm). The quantum dots were characterized using photoluminescence and UV-Vis spectroscopy.

A01604-03986

Development of Iron Oxide Nanoparticle Based Electrochemical Immunosensor for the Detection of Human PSA

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The electrochemical immunosensor has gained considerable attention in protein detection due to several advantages such as sensitivity, rapid analysis, low cost, small volume needed for analysis and low consumption of materials. Prostate-specific antigen (PSA) is a prominent marker for cancer diagnosis. In the present work, development of an ultra sensitive functionalized iron oxide nano particle based amperometric electrochemical sensor was investigated and tested against human PSA. The novelty and significance of the use of iron oxide nanoparticles is that it enables the sensor to act as a transducer as well as an immobilization matrix for the antibody. The amperometric electrochemical cell consists of platinum as an anode, disposable screen printed electrode (SPE) of carbon mixed with iron oxide based enzyme linked by anti PSA film as cathode and SCE as reference electrode.

A low cost, immunosensor was constructed using a plastic sheet as a base, screen printed with a paste of iron oxide and carbon with resin as a binder. The surface of the electrode was coated with anti PSA, enzymes such as biotin and streptavidin and tested against PSA. The electrochemical detection potential was fixed vs. SCE. The electrochemical cell reaction was carried out in phosphate buffer saline as a conducting medium.

Several experimental parameters such as reaction time, immobilization time, concentration of anti -PSA immobilized in iron oxide and other electrochemical parameters were studied by injecting PSA into the electrolyte for detection. Immunochemical reaction produced a maximum current response at pH 7.4 of PBS. Incubation time vs. electrical characteristics was performed; the current response increased linearly with the incubation time upto 10min and later it reached the saturation state. Consistent results were obtained for the enzyme linked iron oxide electrochemical sensor. It was found that the sensor exhibited linear response for PSA concentration in the range from 200 ng /ml to 0.5 $\mu\text{g}/\text{ml}$, after which the response saturated, the corresponding current variation was in the range from 10^{-7} A to 10^{-5} A. Thus this sensor exhibits considerable promise for early detection of cancer; these results will be discussed in detail in the presentation.

A01638-02838

Biological Applications of Colloidal NanoparticlesWolfgang PARAK*Fachbereich Physik, Philipps Universitaet Marburg, Marburg, Germany*

Two ideas for applications in which colloidal nanoparticles are used to interface cells are discussed. For this the report will be structured in three parts. In the first part the colloidal nanoparticles and in particular their surface chemistry will be discussed. In the second part a nanoparticles based FRET system for the detection of intracellular ions will be reported. In the third part an alternative system for the detection of intracellular ions based on polymer capsules / nanoparticles based will be described.

With a very general procedure inorganic hydrophobic nanoparticles of different materials can be transferred into aqueous solution by coating them with an amphiphilic polymer that is based on a poly(maleic anhydride) backbone modified with alkylamine sidechains. Functional groups with an amino anchor can be directly introduced in the polymer by reacting them to anhydride groups of the backbone. This offers a very general route to water-soluble nanoparticles of high colloidal stability, with good size distribution, and with a variety of functional groups that are directly embedded in the polymer shell without the need of post-bioconjugation.

A FRET-pair based on colloidal quantum-dot donors and multiple organic fluorophores as acceptors is reported. In contrast to similar systems which are used as biosensors and detect specific changes of the donor/acceptor-distance under the influence of analyte binding, our nanoparticle design seeks to optimize sensors that detect spectral changes of the acceptor at fixed donor/acceptor distance. This approach allows for relatively small acceptor-donor distances, and thus for high energy transfer efficiencies, while simultaneously permitting high colloidal stability. The photophysics of the system is characterized and compared to similar systems which have been reported in literature.

Polyelectrolyte microcapsules have been loaded with a pH sensitive, high molecular weight SNARF-1-dextran conjugate. SNARF-1 exhibits a significant pH-dependent emission shift from green to red fluorescence under acidic and basic conditions, respectively. The spectral properties of the dye were found to be largely retained after the encapsulation. Upon ingestion of SNARF-1-filled capsules by breast cancer cells or fibroblasts, the pH change of the local capsule environment during transition from the alkaline cell medium to the acidic endosomal/lysosomal compartments could be observed. By incorporating magnetic and fluorescent colloidal nanoparticles into the capsule-shell a novel type of multiplexed sensor system was developed.

A01662-02874

Facile Synthesis of Ag₂O ColloidsLian-Ming LYU; Michael H. HUANG*Department of chemistry, National Tsing Hua University, Hsinchu, Taiwan*

Promising applications of silver oxide (Ag₂O) nanoparticles for optics, catalysis and nanodevices have recently been demonstrated. Ag₂O can be easily prepared by mixing aqueous solution of silver nitrate and an alkali hydroxide. Nevertheless, few reports about synthesizing uniform silver oxide nanocrystals with well-defined shapes were published, let alone information about their applications.

Our laboratory has found a facile direct growth strategy for the synthesis of submicron-sized Ag₂O crystals of different shapes by an injection method. We have changed the solution conditions by varying the concentrations of the reagents, the sequence of their introduction, and temperature to induce the formation of these different morphologies. The particles were formed within seconds. Interestingly, we can easily convert Ag₂O to Ag₂S by adding a sulfuric ion source and some other reagents.

A01687-02916

Routes to Biomedical Applications of Mesoporous SilicaChung-Yuan MOU*Department of Chemistry, National Taiwan University, Taipei, Taiwan*

Our goal is to fully develop the potential of mesoporous silica in biomedical applications. Mesoporous silica nanoparticles (MSN) with unique properties such as high surface area, uniform and tunable pore size, easy modification, and biocompatibility have made them suitable for biological applications. The nanoparticles could serve as a reservoir/carrier for drugs, enzymes, DNA, or as a nano-reactor converting prodrugs into drugs inside cells.

For the past few years, we have synthesized aqueous dispersible, well-ordered, size-controllable MSN with pore size ~3 nm and uniform particle size of 30 to 200 nm. We have attached imaging functionalities, fluorescence and magnetic resonance imaging contrast enhancement, to the nanoparticles. We have demonstrated that the resulting nanoparticles could be uptaken by many different cells, such as 3T3-L1/NIH 3T3 murine fibroblast cells, HeLa, MCF-7, BT474, and most importantly human mesenchymal cells (hMSC), with high efficiency. This high efficiency is a prerequisite for a cell marker. The presence of nanoparticles does not have adverse effect on the cell viability, proliferation, and differentiation on the stem cell hMSC. The magnetic Mag-Dye@MSN and Gd-

Dye@MSN can act as MRI contrast agent and fluorescence tracker as evidenced in animal tests. Targeting capability has also been demonstrated by attaching a monoclonal antibody, Herceptin, to MSN and target to HER2 protein demonstrated.

Three recent results will be highlighted in the lecture:

(1) Near-infrared Mesoporous Silica Nanoparticles for Optical Imaging: Characterization and In Vivo Biodistribution: MSN was adsorbed with indocyanine green (ICG) by electrostatic attraction to render MSN-TA-ICG as an efficient NIR contrast agent for in vivo optical imaging.

(2) Mesoporous Silica Nanoparticles Functionalized with Oxygen-Sensing Probe for Cell Photo-Therapy as Potential Cancer Theranostics: MSNs platform extends the Pd-porphyrin from its renowned function as a phosphorescence probe for oxygen sensing/imaging (diagnostics) to a novel nano-photosensitizer for cell photo-therapy (therapeutics).

(3) Size Control of Well-dispersed, Uniformed Mesoporous Silica Nanoparticles and the Size Effect on Cell Labeling: Well-suspended uniform mesoporous silica nanoparticles (MSNs) were prepared by a simple two-step method. Size-controlled MSNs, ranging from 30 to 280 nm, were obtained by adjusting the pH of the reaction solution. The uptake of FITC-MSNs by HeLa cells is size-dependent with maximum uptake at a nanoparticle size of 50 nm.

A01690-02925

Ultrafast Dynamics Studies of ZnS Semiconductor Nanoparticles

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The optical properties of semiconductor nanoparticles have been under scrutiny for years. Concepts pertaining to particle size, crystal imperfections, temperature dependence and dopant emission have all been invoked to explain the plethora of optical phenomena. However, there has been no coherent model correlating all these disparate concepts, leading to confusion over the numerous reports. Here we present a new picture that accounts for these four concepts, fundamental to the study of semiconductor materials. These are manifested in the localized donor-acceptor-pair (DAP) recombination, and the new mechanisms of phonon-modulated-pairing (PMP) and resonant-energy-transfer (RET). Our recent experiments utilize the wide bandgap ZnS nanoparticles as the prototype system. Contrary to conventional perceptions, the imperfections in nanoparticles are essential to the energy transfer in doped

nanoparticles. Particle size and thermal energy also dictates the nominal DAP energy that is required for RET to the dopant subsystem, yielding enhanced luminescence.

A01750-03052

Size Controllable Supramolecular Nanoparticles for Molecular Diagnostics and Therapeutics

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Over the past decades, there have been significant efforts devoted to explore the use of nanoparticles in the fields of biology and medicine. Several different types of nanoparticles have successfully made their ways into pre-clinical studies, clinic trials or even successful commercial products used in routine clinical practice. However, there remains an imperious desire for developing novel synthetic approaches in order to produce new-generation nanoparticles which have (i) controllable sizes and morphologies, (ii) low toxicity, compatible immunogenicity and *in vivo* degradability, and (iii) proper surface charges and chemistry for improved physiological stability and longer circulation time. Moreover, multiple functions – for example, reporter systems for real-time monitoring with imaging technologies (i.e., optical imaging, magnetic resonance imaging and positron emission tomography), targeting ligands for disease-specific delivery, and controllable mechanism for packaging and releasing of drugs and genes – will be conferred to individual nanoparticles for conducting multiple applications in parallel.

We have recently developed a convenient, flexible and modular synthetic approach for preparation of size-controllable supramolecular nanoparticles (SNPs) from different molecular building blocks. A collection of SNPs with variable sizes ranging from 30 to 450 nm were prepared by simply mixing the molecular building blocks at different ratios, and the resulting SNPs exhibit good stability against the variations of temperatures, pH values and ionic strengths. Two unique dynamic characteristics of the SNPs (i.e., competitive disassembly and reversible size controllability) were examined to further validate the molecular mechanism of this supramolecular approach. To explore biomedical application of the SNPs, we carried out positron emission tomography imaging studies by injecting ⁶⁴Cu-labeled SNPs with different sizes into mice. Both whole-body biodistribution and lymph node drainage studies revealed that the sizes of SNPs affect their *in vivo* characteristics.

It is conceivable that the supramolecular chemistry-based concept demonstrated here can be further explored for producing size-controllable nanostructured materials from a wide range of molecular building blocks, e.g., organic

motifs, metal clusters, DNA/peptide aggregates and water-soluble oligomers/polymer. Besides the imaging studies mentioned above, we are currently exploring the use of the size-controllable SPNs for gene delivery and cancer vaccine. Much extensive and in-depth study will reveal the potentials of these SPNs.

A01759-03088

Synthesis of Nanoparticles of Porous Iron Carboxylates for Biomedical Applications

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Porous Metal-Organic-Frameworks (MOFs) are promising candidates for biomedical applications including drug delivery, controlled delivery of nitric oxide or imaging. In addition, some of them (iron carboxylates) exhibit a low toxicity. However, their uses required often *in vivo* administration and there is a strong need to develop new synthesis routes of MOF nanoparticles, sometimes functionalized with surface agents to ensure a furtivity upon *in vivo* administration.

This work deals with the synthesis of nanoparticles of porous iron (III) carboxylates as new drug delivery or imaging systems, through the use of various synthesis techniques (solvolthermal, microwave, ultrasonic...). Different compositions have been investigated and particles with sizes between 60 and 200 nm have been obtained. Surface modification agents (polymers) have been introduced at different stages of the synthesis. First encapsulation and release tests of cyto-toxic drugs have been also performed indicating a faster release of the drugs when using nanoparticles while their magnetic resonance imaging properties have been investigated, revealing a surprising imaging activity which makes these nanoparticles promising candidates as contrast or theranostic agents.

A01763-03089

Nuclear Magnetic Resonance Investigation of Novel Mn-Ferrites and Co-Ferrites Based MRI Contrast Agents

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The Nuclear Magnetic Resonance Dispersion (NMR-D) profiles of different classes of possible novel contrast agents (CA) for Magnetic Resonance Imaging (MRI) is presented.

The samples consist in Mn-ferrites and Co-ferrites compounds made up of nanoparticles (NP) with different magnetic cores ($Mn_{3-x}Fe_xO_4$ and $Co_{3-x}Fe_xO_4$ respectively). These compounds were obtained by rapid decomposition of metalcarbonyl into a hot solvent and a coordinating surfactant, followed by an oxidation step. In this way we obtained, by controlling the metal/surfactant molar ratio, various samples with different sizes of the monodisperse capped nanoparticles.

We performed several characterization measurements, the first ones headed to a structural and a morphological investigation by XRD and TEM techniques.

Zero-field-cooled and field-cooled SQUID magnetization measurements revealed for all the samples a superparamagnetic behaviour with blocking temperatures in the range 30-150K.

The study of NMR-dispersion profiles revealed r_1 and r_2 relaxivities (i.e. the efficiency in contrasting MR images), for most of our samples, comparable to commercial compounds for frequencies $f > 100$ MHz, resulting in a high efficiency for high-field clinical and research Imagers. Furthermore, the Co-ferrites at low and intermediate frequencies revealed relaxivities higher than commercial SP compounds. The comparison of the relaxivities of the two series of samples allowed to highlight the crucial role of the magnetic anisotropy and of the kind of magnetic ion for the nuclear relaxation mechanism.

A01763-03805

A Novel Approach to Magnetic Field Biosensors: NMR and SQUID DetectionAndrea VALSESIA¹; Pascal COLPO¹; Francois ROSSI¹; Paolo AROSIO²; Manuel MARIANI^{3,4};Alessandro LASCIALFARI^{2,3,4}; Maurizio CORTI^{3,4}

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We have studied a novel approach for the realization of Magnetic Field Effect Biosensors (MFB), by optimizing the technique of immobilization of biomolecular probes on the surface. By using magnetite nanoparticles for marking the biomolecules, we obtained a good sensitivity of the detection method of MFB using nuclear magnetic resonance (NMR) and SQUID. The modification of the surface chemistry of the substrate was obtained by depositing via Plasma Enhanced Chemical Vapour Deposition a 50 nm-thick layer of Poly Acrylic Acid (ppAA), characterized by high surface density of COOH groups. The proof-of-concept of the MFB was designed using the following two protocols: in the "labelled" protocol, a monolayer of IgG molecules was adsorbed on the ppAA surface. The IgG molecular probes were exposed to solutions at different Ab-IgG concentrations. The Ab-IgG molecules were labelled by streptavidin. Finally biotinated modified γ -Fe₂O₃ superparamagnetic nanoparticles were flowed in order to be specifically recognized by the streptavidin-modified Ab-IgG. In the "label-free" protocol IgG coated γ -Fe₂O₃ superparamagnetic nanoparticles were immobilized on the ppAA surface, while Ab-IgG at different concentrations were flowed in order to be specifically recognized.

We report on NMR tests performed using "label-free" and "labelled" biosensors, whereas SQUID measurements have been performed just on labelled sensors. In NMR experiments a change in ¹H nuclear relaxation rates due to the presence of magnetic nanoparticles was observed, while in SQUID measurements the system passes from being diamagnetic to a typical paramagnetic behaviour in presence of magnetite nanoparticles.

A01820-03169

A Proteomic Characterization of a Golgi-associated Bi-lobed Structure in Trypanosoma BruceiQing ZHOU; Cynthia Y. HE

Department of Biological Sciences, National University of Singapore, Singapore

Trypanosoma brucei is a pathogenic protozoan that causes human sleeping sickness in sub-Saharan Africa. Its highly simplified cellular organization makes it useful as a model organism to study the molecular mechanism of organelle duplication during the cell division cycle. A bi-lobed structure was recently discovered that is required for duplication of the Golgi apparatus as well as efficient cell division. Several proteins important for cell cycle regulation such as centrins (small calcium-binding proteins mostly found associated with centrosomes) and a polo-like kinase have been found present on the bi-lobed structure, suggesting an important role of this structure in the control of cell cycle progress in *T. brucei*. To further characterize this novel structure and to understand its biological functions, we performed comparative proteomics analyses on partially purified bi-lobe fractions. New bi-lobe proteins were identified, and their localizations were analyzed by fluorescent protein tagging.

A01847-03203

Synthesis, Properties and Assembly of Complex Nanocrystal StructuresLiberato MANNA

Fondazione Istituto Italiano di Tecnologia, Genova, Italy

Current efforts and success of nanoscale science and technology are related to the fabrication of functional materials and devices in which the individual units and their spatial arrangement are engineered down to the nanometer level. One promising way of achieving this goal is by assembling of colloidal inorganic nanocrystals as the novel building blocks of matter. This trend has been stimulated by significant advancement in the wet-chemical syntheses of robust and easily processable nanocrystals in a wide range of sizes and shapes. The increase in the degree of structural complexity of solution-grown nanostructures appears to be one of the natural directions towards which nanoscience will increasingly orient. Recently, several groups have indeed devised innovative syntheses of nanocrystals through which they have been able to group inorganic materials with different properties in the same particle. These approaches are paving the way to the development of nanosized objects able to perform multiple technological tasks. This talk will review the recent advances in the synthesis of colloidal nanocrystals,

with emphasis on the strategies developed in our labs for the fabrication of colloidal nano-heterostructures, as well as on their properties and their assembly.

A01877-03238

Novel Functionalized and Nanostructured Conducting Polymers for Biosensing

Jadranka TRAVAS-SEJDIC

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The great demand for fast, label-free gene detection has prompted extensive research into alternative approaches to traditional DNA assays that employ a range of readout modalities, including optical, acoustic and electronic methods. Promising approaches utilize novel smart and nanostructured materials.

We have developed a number of novel DNA sensors that are based on conducting polymers (CPs) and allow rapid one-step detection of unlabeled DNA fragments. These sensors are based on functionalized and nanostructured conducting polymers derived from polypyrrole, polythiophene and polyaniline. Such CPs sensors provide intrinsic electrical readout using convenient electrochemical techniques. Complementary to such solid substrate sensors we have also demonstrated a novel solution-based DNA assay with optical readout based on a new poly(p-phenylenevinylene)-derived cationic conjugated polymer.

In this talk these novel gene sensing approaches will be discussed, as well as our recent results on development of nanostructured CPs for utilization in sensing and biosensing.

A01886-03256

An Air-Supported Liquid Crystal System for Real-Time and Label-Free Characterizations of Phospholipases and Their Inhibitors

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Lin-Yue Lanry YUNG

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A new air-supported liquid crystal (LC) system for analyzing interfacial phenomena that occur based on the molecular interaction between LCs and adsorbed molecules of interest at the aqueous/LC interface is reported. Compared with existing LC-based detection systems, the miniature system reported here requires less sample and involves simpler preparation. Using this system, the enzymatic hydrolysis of various phospholipases such as phospholipase A2 (PLA2), phospholipase C (PLC) and phospholipase D (PLD) are characterized. The hydrolysis

of phospholipid monolayers self-assembled at aqueous/LC interface induces an orientational response from the LCs. As a result, an optical signal that reflects the spatial and temporal distribution of phospholipids during the enzymatic reaction can be generated in a real-time manner. When well-known phospholipase inhibitors are introduced together with respective phospholipases, no orientational response of LCs is observed. In the case of inhibitors MJ33 and compound 48/80, cross-inhibitions among phospholipases are also observed. This work demonstrates that the air-supported LC system provides a facile label-free assay for characterizing phospholipase activities and for screening enzyme inhibitors. It could potentially be useful for different high throughput and cost-effective enzyme screening assays.

A01893-03263

Selective Growth of Gold Nanoparticles onto Tellurium Nanowires via a Green Chemical Route

Zong-Hong LIN; Yang-Wei LIN; Kun-Hong LEE;
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In this study, we describe a green chemical route for highly selective growth of Au tips onto Te nanowires. We carefully selected the pH values to vary the redox reaction potential between Au^{3+} and Te nanowires, allowing control of nucleation and growth rates of Au nanoparticles. In the presence of 10 mM CTAB, we obtained Au-Te and Au-Te-Au nanowires at pH 4.0 and 5.0, respectively. Photovoltaic data revealed that the resistance of the Te nanowires is controlled by the degree of deposition of Au nanoparticles. We suspect that Au-Te and/or Au-Te-Au nanowires hold great potential for use in the fabrication of electronic devices. In the other way, through strong Au-S bonding, the as prepared Au-Te nanowires were subjected to assemble in the presence of 1,6-hexanedithiol or 1,4-benzenedithiol. We further demonstrated that the self-assembled Au-Te nanowires each having a larger Au nanoparticle provide a greater surface-enhanced Raman scattering (SERS) enhancement (1.8×10^6 vs. 5.6×10^4 fold compared to that without using a matrix) in the presence of 1,4-benzenedithiol.

This study was supported by the National Science Council of Taiwan under contracts 96-2627-M-002-013 and 96-2627-M-002-014.

A01895-03268**Synthesis and Characterization of $Zn_xHg_{1-x}Se_yS_{1-y}$ Quantum Dots**Guo-Yu LAN; Yang-Wei LIN; Zong-Hong LIN;
Huan-Tsung CHANG*Department of Chemistry, National Taiwan University,
Taipei, Taiwan*

In this research, we describes the synthesis of highly water-soluble $Zn_xHg_{1-x}Se_yS_{1-y}$ quantum dots (QDs) in aqueous solution through a simple photo-assisted reaction between ZnSe QDs and mercury(I) nitrate dihydrate [$Hg_2(NO_3)_2 \cdot 2H_2O$]. To deduce the optimal synthesis conditions, we varied several parameters, including the concentrations of mercaptosuccinic acid (MSA) and $Hg_2(NO_3)_2 \cdot 2H_2O$, the illumination time, and the reaction temperature. When irradiated at temperatures below 80°C, the ZnSe QDs reacted with the S^{2-} ions formed rapidly from MSA and the Hg^{2+} ions formed from Hg_2^{2+} ions to form $Zn_xHg_{1-x}Se_yS_{1-y}$ QDs through a process of photo-etching and surface combination. Under different conditions, we prepared a series of $Zn_xHg_{1-x}Se_yS_{1-y}$ QDs that emit fluorescence at the maximum wavelengths ranging from 405 to 760 nm. Inductively coupled plasma-mass spectrometry (ICP-MS) and scanning electron microscopy/energy dispersive spectrometry (SEM-EDS) revealed that the content of Hg in the $Zn_xHg_{1-x}Se_yS_{1-y}$ QDs was greater when the synthesis was conducted at higher temperature. The $Zn_{0.60}Hg_{0.40}Se_{0.50}S_{0.50}$ QDs exhibit improved photostability than crude ZnSe QDs and possess long lifetimes ($\tau_1 \sim 38$ ns and $\tau_2 \sim 158$ ns).

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A01896-03269**Fabrication of Efficient CdHgTe/CdTe Quantum Dots Sensitized Solar Cells**

Zusing YANG; Huan-Tsung CHANG

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We have demonstrated fabrication of quantum dot-sensitized solar cells (QDSSCs) using CdHgTe nanocrystals (NCs) and CdTe quantum dots (QDs). The two nanomaterials were separately prepared from aqueous mixtures of NaHTe, $Cd(NO_3)_2$, and 3-mercaptopropionic acid in the absence and presence of $HgCl_2$. The absorbance values of the CdHgTe NCs over the wavelength range from 400-1200 nm are higher than those of CdTe QDs. They were deposited onto TiO_2 electrodes through a self assembly technique and then the electrodes were used to prepare $(CdHgTe)_3$ and $(CdHgTe)_3-(CdTe)_2$ QDSSCs.

The energy conversion efficiency (η) values of the two QDSSCs are 1.0% and 2.2%, respectively. The incident photon to current conversion efficiency (IPCE) data reveal that $(CdHgTe)_3$ and $(CdHgTe)_3-(CdTe)_2$ QDSSCs have 39% and 66% at 400 nm, respectively, and both have 16% at 800 nm. Our present results show the possibility of fabricating higher efficient QDSSCs using CdHgTe NCs and other nanomaterials that provide greater absorption coefficients and luminescent quantum yields.

Acknowledgments:

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A01998-03448**Ultra-Sensitive Cholesterol Biosensor Based on ZnO Nanoparticles Grown in Solution**Sang Hoon KIM; Mohammad VASEEM; Ahmad UMAR;
Yoon Bong HAHN*School of Semiconductor and Chemical Engineering,
Chonbuk National University, Jeonju, South Korea*

A high-sensitive amperometric biosensor based on the immobilization of cholesterol oxidase (ChOx) onto the ZnO nanoparticles has been fabricated which shows a very high and reproducible sensitivity of $23.7 \mu A mM^{-1} cm^{-2}$, detection limit (based on S/N ratio) 0.37 ± 0.02 nM, response time less than 5 s, linear range from 1.0 ~ 500.0 nM and correlation coefficient of $R = 0.9975$. A relatively low value of enzyme's kinetic parameter (Michaelis-Menten constant) ~ 4.7 mM has been obtained which indicates the enhanced enzymatic affinity of ChOx to Cholesterol. To the best of our knowledge, this is the first report in which such a very high-sensitivity and low detection limit has been achieved for the cholesterol biosensor by using ZnO nanostructures modified electrodes.

A02068-03558

SERS Detection of DNA at Sub-Picomolar Level on Silicon Nanotips ArraysHsin-I HSIUNG¹; Hung-Chun LO²; Li-Chyong CHEN¹; Kuei-Hsien CHEN²*1. Center for Condensed Matter Sciences, National Taiwan University, Taipei, Taiwan**2. Institute of Atomic and Molecular Sciences, Academia Sinica, Taipei, Taiwan*

Surface-enhanced Raman spectroscopy (SERS) is a well-established hassle-free less-destructive and highly sensitive technique for the detection of fluorescent biomarkers or nonfluorescent biospecies like DNA. Literature reveals that the SERS detection of DNA can achieve the sensitivity at as low as 10^{-13} M of concentration, by employing the dye labeled-DNA dispersed in silver nanoparticles (AgNPs) containing buffer solution, however it is an indirect detection in solution phase and consequently suffers from the foul-up (contamination) and complication in process, and hence results in unstable and unreliable SERS signals in comparison with the direct detection of the analytes on the AgNPs decorated solid substrates.

On the other hand, during the present decade, the wafer-scale silicon nanotips arrays (SiNTs), fabricated by a patented self masked dry etching technique, has established as an excellent platform for the self assembly of metal NPs. The hybrid of metal NPs and SiNTs have exhibited an excellent performance as a template for SERS detection for detection and identification of the molecules like Rhodamine 6G and trans-1,2-bis(4-phyridyl)ethylene (BPE), with the enhancements in the range of 10^6 to 10^8 , in the concentrations range of 10^{-6} to 10^{-10} M. Employing the semiconductor compatible process like electron cyclotron resonance microwave plasma enhanced chemical vapor deposition (ECR-MWPECVD), the well aligned SiNTs are fabricated, from the 4 inch single crystalline silicon wafer, under the ECR plasma of silane (SiH_4), methane (CH_4), hydrogen (H_2) and argon (Ar) gas mixture. These dry substrates possess remarkable stability and reproducibility in comparison with those prepared by wet chemistry.

In our study, the thiol-functionalized 20-base double-strand DNA sequences of adenine (A) and thymine (T) were chosen for AgNPs-assisted SERS detection. However, the poor affinity of Ag with thiol could not provide stable immobilization of DNA on pristine SiNTs. Therefore, prior to the AgNPs decoration, Au was coated on the SiNTs by ion beam sputtering deposition (IBSD) leading to the formation of well aligned arrays of AuNTs (Au-coated SiNTs) with apex diameter of ~ 10 nm, bottom diameter of ~ 200 nm, length of ~ 1600 nm and density of 10^{11} cm^{-2} . The size of AgNPs varied 3-10 nm in diameter, and the inter-particle distance is about 1-2 nm.

A high aspect ratio AuNTs decorated by AgNPs exhibit a proper DNA-immobilization via strong Au-S bonds, and hence a highly sensitive DNA detection via SERS technique. Ultra low DNA concentration, about 10^{-13} mol cm^{-3} , can clearly demonstrate an enhanced and stable Raman signal. The DNA-immobilized substrates possess excellent stability and reproducibility: Raman signals retain the activity even after six months conservation in room temperature. The relatively simple, binder-less, stable, room temperature and label-free bio-molecular detection underlines the effectiveness of these substrates for other bio-molecular systems.

A02080-03589

Highly Sensitive and Selective Detection of Mercury Ions by Using Oligonucleotides, DNA Intercalators and Conjugated Polymers

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Conjugated polymers have been known to display interesting optical properties such as optical amplification effect via fluorescence resonance energy transfer (FRET). They can thus be used as the optical platforms for highly sensitive chemical and biological sensors. Here we demonstrated a practical scheme for the detection of mercury ions in aqueous media at room temperature with high sensitivity and selectivity by using a combination of oligonucleotides, DNA intercalators and conjugated polymers. This scheme combines the advantages of specific binding interactions between Hg^{2+} and thymine, and optical amplification properties of conjugated polymers. This method is label free, low cost and simple to use, and all of the materials are commercially available. It works in a "mix-and-detect" manner. The limit of detection could be improved to 0.27 nM, which is much lower than the maximum level of mercury permitted by the EPA in drinking water. This scheme could also be potentially used as a two-photon sensor for detecting mercury ions in a biological environment where deep penetration is required. A detection limit of as low as ~ 6 nM could be achieved under two-photon excitation.

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A02190-03749

Gold Nanoparticles for Immuno Sensing using Darkfield Imaging

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Gold nanoparticles (AuNPs) experience a shift of their extinction maximum upon changes in the refractive index of the material surrounding them. We have used this effect to construct an immunosensor using a darkfield microscope equipped with a colour camera.

The shift of the extinction maximum of AuNPs scales with the change of refractive index of the medium surrounding them, which in turn is determined by the composition of this surrounding material. The larger the change in surrounding material the larger is the change in refractive index and, therefore, also the change of the extinction maximum, thus providing a method to study binding events on AuNPs by observing the induced changes of their extinction spectra, i.e. their change of colour.

To this end, we thiolated various antibodies using iminothiolane and subsequently conjugated them to uncapped AuNPs. The conjugation reaction could be shown to be tunable regarding the amount of adsorbed material, which is proportional to the resulting change of colour. Furthermore, the amount of active antibody on the surface of the AuNPs could be determined using a fluorescently labelled antigen. We compare these results with discrete dipole simulations of the expected colour change as a function of adsorbed protein. We then immobilised AuNP conjugates in micro flow cells and studied them using spectrally-resolved darkfield imaging microscopy. Upon immunoreaction a change of colour could be observed proving the viability of this setup as a nanoparticle-based immunosensor.

A02202-03780

Fabrication of Chemical and Biosensors Based on ZnO Nanostructures by Electrochemical Principle

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The diversity in the properties of ZnO makes it a versatile material for the wide ranging application possibilities which provide it an opportunity to presents itself as one of the most promising material for the future research and applications. The nanostructures of ZnO are of particular interest as they combine different properties such as high specific surface area, optical transparency, biocompatibility, non-toxicity, chemical and photochemical stability, ease of fabrication, and so on. Even having versatile properties, the chemical and biosensor applications of ZnO nanostructures are rare. Here, we present the synthesis and chemical and biosensors applications of well-crystallized zinc oxide nanonails grown via the thermal evaporation process. The detailed structural and optical characterizations confirmed that the as-synthesized nanonails are single-crystalline with the wurtzite hexagonal phase, grown along the [0001] direction and possessing good optical properties. A high sensitive with low detection limit glucose biosensors based on the glucose oxidase immobilized on ZnO nanonails and hydrazine chemical sensors have been presented here. The ZnO nanonails were used as supporting matrixes for enzyme immobilization, glucose oxidase (GOx), to construct efficient glucose biosensors. As the ZnO nanonails have a high surface area and presenting themselves as an efficient electron conducting tunnel. Therefore, GOx attached to the surfaces of ZnO nanonails had more spatial freedom in its orientation, which facilitated the direct electron transfer between the active sites of immobilized GOx and electrode surface. A high sensitivity, $24.613 \mu\text{A cm}^{-2} \text{mM}^{-1}$, with a response time less than 10s was achieved from the fabricated glucose biosensor. The biosensor shows a linear range from 0.1 to 7.1 mM with a correlation coefficient of $R= 0.9937$ and the detection limit of $5 \mu\text{M}$. The detection of hydrazine was performed in a phosphate buffer (pH 6.6). The peak current increases linearly with the concentration of hydrazine from 0.01 to 1mM. This work demonstrates that the ZnO nanostructures can be utilized as an efficient electron mediator to fabricate efficient chemical and biosensors.

A02210-03989

A02221-03797

Enhancing the Sensitivity of Silicon Nanowire Bio-Chemical Sensor

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Nanotechnology is being used to create many new materials and devices with wide-ranging applications such as in medicine, electronics, and energy production which are impacting our daily life. With the increasing spread of knowledge, healthcare has been brought into focus which includes early disease diagnosis, drugs discovery, etc. For these applications specific and sensitive detection of various biological and chemical species becomes crucial. Among various nano-materials, nanowire (NW) based bio-chemical sensors are most exploited for the purpose. They are realized using chemical vapour deposition (CVD) and standard silicon processing technologies. The NW sensors work on the principle of Field Effect Transistor (FET) where charge associated with the chemical species attached on the nanowire surface acts as the chemical gate, hence are also termed as CHEM-FET.

We realized N-type silicon nanowires in array format, with cross-sectional dimensions ~50nm and length 100µm using CMOS compatible silicon processes. The device have thin silicon oxide cover on nanowires as the gate dielectric. The two ends of Si NW are integrated with silicon pads which are connected to metallic inter-connects and pads that allow interface to macroscopic instruments. The chips can be built with integrated fluidic channels or open reservoirs for the easy dispensing of analytes. We tested Phosphate buffered saline (PBS) with different pH as varied gate potential and their effect on the nanowires current was recorded. The effect of back gate potential (V_g) to modulate the sensitivity of the sensor was studied.

Compared to neutral (pH 7.4) PBS, when an acidic PBS (pH 4.1) contacts SiNW, it's current showed about 23% and no change at biasing potential of -0.5V and 0.5V respectively. The sensor current change was significantly enhanced, by the application of back-gate bias (V_g = -1V), to 48% and 29% at two NW biasing conditions, enhancing the nanowire sensitivity. For the alkaline PBS (pH 8.7) similar trends were also observed and the nanowire current changes more than 40% were observed. Detailed experimental set-up, results and their analysis will be presented. Statistical data from multiple sensors in a chip and from multiple chips will also be elaborated.

Bioconjugated Nanostructures for Diagnosis and Therapy

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Magnetic nanoparticles are being studied because of their numerous applications in the biomedical field as magnetic contrast agents in magnetic resonance imaging (MRI), colloidal mediators for cancer magnetic hyperthermia and vectors that can be directed by means of an external magnetic field towards a desired location, such as in the case of the targeted drug delivery.

Incorporation of optical functionality to magnetic nanoparticles is of growing interest. The combination of optical and magnetic properties in a single nanoprobe would allow simultaneous biolabeling/optical or magnetic imaging and cell sorting/separation, which could be of enormous importance, both from a diagnostic and therapeutic point of view. The interest in magneto-optical nanoparticles is still higher if the particles are water soluble, a requisite for biomedical applications.

Ferritin is an example of a genuine magnetic bionanoparticle. Ferritin is the major intracellular storage form of iron and is a spherical protein capable of accommodating an iron oxide mineral. Recently, we have reported that native ferritins are basically composed of a ferrihydrite-magnetite core-shell structure. Specifically, magnetite appears to be the predominant phase when ferritin iron content is gradually removed, reaching a ~70% in magnetite for the 200 Fe ferritin (Magnetoferritin). A high spontaneous magnetization value of 70 emu/gFe was obtained (M_s=95 emu/gFe for magnetite). Furthermore, the exterior shell of ferritin provides a platform for chemical and genetic functionalization. In fact, we have lately demonstrated that ferritin can be simultaneously conjugated with appropriate different Alexa Fluor dyes exhibiting fluorescence resonance energy transfer (FRET) in a single nanoparticle.

Semiconductor nanoparticles or quantum dots (QD) have emerged as an important alternative to "classic" organic dyes. In comparison with organic dyes and fluorescent proteins, QDs have unique optical and electronic properties: high photostability, high emission quantum yield, narrow emission peaks, size-dependent wavelength tunability and molar extinction coefficients 10–50 times larger, which make them much brighter in vivo conditions.

Diverse application possibilities, including fluorescent labels for optoelectronics and for biology (biolabeling and bioimaging) are currently being pursued.

A new class of water soluble fluorescent-magnetic Quantum Dots-Magnetoferritin bioconjugate was prepared. The nanostructures were patterned mainly as dimer particles as characterized by High Angle Annular Dark Field Scanning Electron Microscopy (HAADF-STEM) and Electron Energy Loss Spectroscopy (EELS). Magnetic (high spontaneous magnetization values, superparamagnetism) and fluorescent (narrow emission peaks, uniform brightness) properties of both nanoblocks remain in the final nanostructure.

A02230-04476

A GMR Sensor- and High-moment Magnetic Nanoparticle-based Biomedical Sensing System of Zeptomol (10^{-21} mol) Sensitivity: An Integrated Platform Potentially Leading to Personalized Medicine

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Longitudinally monitoring the changes of protein biomarkers (generally < pmol) is expected to help design medical treatment for specific individuals and detect chronic diseases in their early stage, which is critical to achieve successful personalized medicine and control of chronic diseases. This requires a highly sensitive detecting system that is of low-cost, easy to use, does not require expensive instruments, and preferentially compatible with current electronic technologies. Since the late 1990s, magnetolectronics, including giant magnetoresistive (GMR) sensor, has emerged as one promising platform to meet such requirements. Several studies have demonstrated the usage of magnetic biosensors to detect biomolecules with relatively large particle labels (> 50nm). The high mass and size of the label in relation to biomolecules to be tethered is expected to interfere with the natural biomolecule movement, recognition, and binding. However, magnetic nanoparticles as detectable labels acquire a smaller magnetic moment than the larger ones, requiring more sensitive sensors and measurement systems. Use of small high-moment magnetic nanoparticles for sensitive detection and quantification of biomolecule are yet to be realized.

In this talk, first we will report the world first development of an integrated magnetic sensing system with GMR sensor-arrays that are several times sensitive than

traditional design, and high-magnetic-moment cubic FeCo nanoparticles that are ten times higher in magnetic moment than commercial iron oxide nanoparticles with same size at 10 Oe field. Then, we will report, for the first time, to detect as few as 600 copies of streptavidin, which is modified by 12.8nm cubic FeCo nanoparticle in a 1:1 ratio. We evaluated the usage of this system to detect human interleukin-6 (IL-6, a potential serum biomarker for lung cancer) using a sandwich approach, which follows the same principle of Enzyme-Linked ImmunoSorbent assay (ELISA). We demonstrated that our magnetic detecting system can adopt the principle of ELISA assay with much increased sensitivity. More importantly, there is a linear dose-response relationship between the amount of IL-6 applied and the magnetic signals detected by the detecting system. GMR and magnetic nanoparticle based detecting system is expected to be applicable to many other biological systems for detection and quantification of various biomolecules. The high sensitivity of this detecting system open new avenues for the detection of biomolecules involved in the etiology of various diseases, especially the chronic ones, such as cancer, which is now under investigation. It can also be used to monitor residual disease and disease reoccurrence. Most importantly, because of the magnetic/electric nature of this detecting system, we expect that this detecting system will help the realization of point-of-care.

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A02231-03810

Creation of Multi-Functional Biointerface Using by Plasma-Processing

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Biomedical polymers such as artificial vessel have contact with tissue, cells and any other biological substances. In this way, the wettability control of the polymers plays an important role because this can control the biocompatibility and the adhesiveness of the surface. We attempted to develop hydrophilic modification used by plasma-initiated polymerization with acrylic acid (AA) and (2-hydroxyethyl) methacrylate (HEMA) and survey the cell growth on such modified polymer surface, and described the regioselective immobilization methods of bacteria.

A02275-03882

Fluorescent Quantum Dot Labels for Rapid and Facile Immunodetection of Multiple ProteinsSonia GAWANDE¹; Anup KALE²; Haribhau GHOLAP^{2,3}; Padma SHASTRY¹; Satishchandra OGALE²

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Nanoparticles possess versatile optical and electronic properties that have wide array of applications in immunoassays. Amongst various nanoparticles, quantum dots have shown great potential in immunodetection, biolabelling and imaging due to unique properties like tunable optical properties, photostability, broad excitation spectra, robustness, narrowband emission, and versatility in surface modification. Quantum dot fluorescence is intrinsic, bright and prolonged compared to the organic dyes and proteins. All these properties make them ideal candidates for interfacing with biomacromolecules. Western blot analysis is a widely used technique in biological research for detection and analysis of specific proteins. However, simultaneous detection of multiple proteins remains a challenge with currently used fluorescent labels. For detecting different proteins, the blot is usually stripped and reprobed with another specific antibody. This approach is time consuming and laborious and may lead to the loss of immobilised proteins from the blot. This problem can be overcome by using quantum dots. Prostate apoptosis response-4 (Par-4) and Poly (ADP-ribose) polymerases (PARP) are indicators of dying cells. Par-4 is a 38 kDa pro-apoptotic protein, expressed at higher levels in cells undergoing apoptosis following calcium ionophores and chemotherapeutic agents in cancer cells. Whereas, PARP-1 is a 115 kDa protein, whose specific cleavage by caspase-3/-7 generates two fragments of 24 kDa and an 89 kDa. This phenomenon has been used generally as a biochemical marker of apoptosis. The simultaneous detection of these two proteins on a single blot is possible by conjugating their specific antibodies with two quantum dots emitting different colors. The antibody for Par-4 is conjugated with CdTe quantum dot emitting green color ($\lambda = 550$ nm) and antibody for PARP-1 is conjugated with CdTe/ZnS core/shell quantum dot emitting orange color ($\lambda = 620$ nm). By using this approach, both proteins, Par-4 and PARP-1 can be easily visualized on the same blot.

A02275-03925

Understanding Nanoparticle-biomolecule Interactions by Fluorescence and Raman SpectroscopyAnup KALE¹; Haribhau GHOLAP^{1,2}; Satishchandra OGALE¹

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CdTe nanoparticles (NPs) capped with Mercaptopropionic Acid (MPA) were synthesized by organometallic route. The freshly prepared NaHTe (as Te precursor) and CdCl₂ were used as precursors. The molar ratio of Cd : Te : MPA was 1:0.2:2.4. The solution was then refluxed under nitrogen flow at 100°C. Synthesis of nanoparticles was confirmed after characterization by UV-VIS spectroscopy, x-ray diffraction (XRD) and transmission electron microscopy (TEM). Highly luminescent nanoparticles with luminescence emission at 540 nm were synthesized. These were subjected to the studies of their interaction with biomacromolecules, a rapidly developing subject. The association of NPs with BMM can influence the properties of both the NPs and the BMM. In our work the interactions between NPs, serum albumin and flavonoids (e.g. Quercetin, Curcumin, Rutin and Naringenin) were investigated by fluorescence and Raman spectroscopy at pH 7.4. Flavonoids are potent naturally occurring therapeutic molecules. Understanding the mechanism of interaction between NPs and serum albumins, is important to develop nanoparticles as light emitting devices, labeling agents and biosensors. On the basis of fluorescence quenching/enhancement results, Stern-Volmer quenching constant (K_{sv}), binding constant (K_a) and binding sites (n) for proteins and flavonoids were calculated. The binding affinities between proteins and different flavonoids were studied. Interaction with BSA enhanced the luminescence of CdTe NPs whereas Hemoglobin and lysozyme quenched it. The results of these studies will be presented and discussed.

A02282-03896

Fluorescent Rare-earth Based Oxides Nanomaterials: From Nanocrystals to Nanorods and Cytotoxicity StudyTimothy Thatt Yang TAN*School of Chemical and Biomedical Engineering,
Nanyang Technological University, Singapore*

Fluorescent rare-earth (RE) based nanomaterials have been extensively studied for potential biomedical application, especially as probes for molecular diagnosis. RE nanomaterials have unique ladder-like energy levels in their shielded 4f orbital, giving rise to unique emission properties such as narrow emission and long emission lifetime. Some rare-earth ions such as gadolinium and dysprosium ions also exhibit paramagnetic properties. The controlled synthesis of RE based nanomaterials with desired phase, composition, morphology, and tailored surface properties will continue to attract widespread research interests in biomedical field. However, there are limited studies on the investigation of the cytotoxicity of the RE nanomaterials. A detail cytotoxicity study of such material is necessary if they were to be applied as diagnostic probes.

This presentation reports a general method of fabricating RE based fluorescent nanocrystals and nanorods with controllable shapes for some RE-oxides including yttrium oxide (Y_2O_3), gadolinium oxide (Gd_2O_3) and dysprosium oxide (Dy_2O_3). The nanorod formation is suggested to be due to self-assembly of nanocrystals in the presence of long-chain alkyl amine as stabilizing agents. The spherical nanoparticles and nanorods can be selectively prepared by varying the synthesis parameters including the reaction temperature, reaction time, and the ratio of yttrium precursors to amine. A detail cytotoxicity study using a new approach based on American National Standard ISO 10993-5 is also presented.

A02296-03920

Homogenous, Highly Emissive QDs-Silica Core-Shell Nanocomposite from Visible to Near Infrared; Their Applications in Two-Photon Imaging of Human Mesenchymal Stem CellsPi-Tai CHOU; Chih-Wei LAI*Department of Chemistry, National Taiwan University,
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In this article, we present the exploration of a synthetic tactic incorporating delay-photooxidation to recover the loss in emission frequently encountered after encapsulating quantum dots (QDs) inside silica shell. This facile synthesis procedure reproducibly increases emissive intensity of QDs (core)/SiO₂ (shell) (60nm) nanomaterials by > 5 folds (QY from 3% to > 15%). The resulting QDs

(core)/SiO₂ proved to be single quantum dot in single SiO₂, homogeneous and highly monodispersed; their emissions have been successfully fine-tuned from visible to near infrared region. We then demonstrate their power in biological imaging by labeling human mesenchymal stem cells under a two-photon confocal microscopy. The results of low cytotoxicity, efficient labeling, and specific location nearby the nucleus characters of these nanoparticles should spark an intensive relevant research within a living system.

A02313-03960

15-Crown-5 Functionalized Au Nanoparticles Synthesized via Single Molecule Exchange on Silica Nanoparticles: Its Application to Probe 15-Crown-5/K⁺/15-Crown-5 "Sandwiches" as Linking MechanismsMei-Lin HO; Yu-Chun CHEN; Pi-Tai CHOU*Department of Chemistry, National Taiwan University,
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Single 15-crown-5 functionalized Au nanoparticles (NPs) were synthesized with assistance of silica particles. The use of silica particles renders intrinsic advantages in that each step along the product formation can be monitored by, for example, a transmission electron microscope, and large surface-to-volume ratio of silica particles leads to a better yield of Au NP attachment than the glass substrate. The as prepared Au NPs, providing access to fundamental analysis, were readily applied in K⁺ recognition and proved to be free from aggregation. Accordingly, a sandwich type of complexation is resolved and the corresponding thermodynamics can be extracted.

A02464-04211

Detection of WSSV Virus with Differential Pulse Voltammetry Technique on Thin ITO FilmSuthisa LEASEN^{1,2}; Somsak DANGTIP^{1,2};
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An electrochemical sensor on transparent conducting electrode such as ITO can offer a simultaneous detection for optical and electrical changes with low background current and good biocompatibility. Thin ITO film was first modified 3-mercaptopropionic acid (MPA). The MPA modified surface was observed to; i) show its absorbance peak at 281 nm in a UV-visible spectrum, ii) exhibit the characteristic sulfur peak under an X-ray fluorescence measurement, and iii) have its roughness decreased from

16.92 nm to 10.33 nm with AFM in comparison with original ITO surface. The 447 base-pair (bp) long ssDNA of white spot syndrome virus (WSSV) was then used as a probe to immobilize on the modified ITO surface. Methylene blue (MB) has consequently been added to check a binding of the probe ssDNA. The oxidation peak of MB at -0.41 V vs Ag/AgCl reference electrode was clearly observed with the differential pulse voltammetry technique. The copious amount of target DNA of 1447 bp long was used for hybridization. After hybridization, the 0.3 M NaCl/15 mM sodium citrate (2xSSC) at 37°C, 1xSSC and 0.5 xSSC at 42°C were used to remove the excessive and non-hybridized strands. The current was observed to be 0.75 and 2.88 μ A for the concentration of 1 pM and 1 nM, respectively.

A02489-04270

The Synthesis and Characterization of QD Incorporated Nano-hybrid Materials

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The stable and brighter fluorescent probes are of considerable interest because of their utilization in ultrasensitive chemo, bio sensor applications detecting toxic or bio-hazard materials.

Here, we have synthesized and characterized nano-hybrid materials incorporated quantum dots and molecular receptors binding toxic metal ions. The hybrid materials have synthesized by the ex-situ and in-situ incorporation of quantum dots into polymer or silica matrix. The polymeric type receptor including the linking group and ion-detecting group was synthesized by the free radical polymerization. High resolution TEM, UV and fluorescence spectroscopy suggest that the nano-hybrids be used for ultrasensitive sensor applications.

A02495-04265

Synthesis and Characterization of Rare Earth Nano Compounds for Bio Labeling Applications

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The nano structures of rare earth compound cerium hydroxide were synthesized. These nano particles were prepared by hydrothermal method, a subsidiary of wet chemical preparatory techniques. High purity cerium oxide was the starting material. Sodium hydroxide was used as precipitating agent. The mixture was hydrothermally treated

at high temperature for overnight period. Washing was done with de-ionized water. Temperature, pH and crystal structure were influencing parameters in nanostructured growth. Structural characterization was done by X-ray Diffraction (XRD) and Scanning Electron Microscopy (SEM). XRD results revealed that the dominating phase is of hexagonal Ce(OH)₃. The particle size estimated by XRD data were in the range 13-20 nm. The synthesized materials have potent applications in bio sensing and bio labeling.

A02533-04334

Synthesis and Characterization of MWNT-ZnO Nanoparticle Composite

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Carbon nanotubes (CNTs) exhibit unique mechanical and electrical properties because of their sp² hybridized nanostructure. Their one dimensional nature and high aspect ratio makes them a special material in order to use them in different microelectronic device application. On the other hand, ZnO is a wide band gap semiconductor having broad range of applications in room temperature ultra violet lasers, sensors, photo-voltaic and nanoelectronic devices. The combination of these two important nanostructured materials has recently attracted interest because they exhibit unique optical and electrical properties.

We have synthesized ZnO-CNT composites by solution method. Multi wall carbon nanotubes are prepared by thermally assisted chemical vapor deposition (CVD) method. The aspect ratio of as prepared CNTs is ~ 4000 (quite high). They are functionalized by the treatment with concentrated nitric acid (HNO₃) as confirmed by IR spectrum. The functionalized multi-walled carbon nanotubes are dispersed in methanol under ultra-sonic agitation. Required amount of zinc acetate is dissolved in methanol to get desired concentration which is added to the CNTs solution. A solution of sodium hydroxide (NaOH) is added drop wise to the above mixture and sonicated for some time. After a couple of hours, the solvent is evaporated and the sample is washed with de-ionized water number of times. The decoration of ZnO particles onto the surface of the CNTs is seen under Scanning electron microscope (SEM). X-ray diffraction (XRD) as a standard technique is employed for structural characterization of the samples. TEM is been used for detailed study of morphology and crystallinity. ZnO nanoparticles are well dispersed on the outer walls of MWNTs.

A02558-04372

Imaging Endogenous Metabolites Directly in Latent Fingerprints Using Silica Nanoparticles and SALDI-TOF Mass SpectrometryAngelina LIM¹; Matthew BENTON³;
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It has been demonstrated that latent fingerprints can be conveniently developed using porous, hydrophobic silica nanopowder to provide excellent definition and resolution of print minutiae. In addition, These silica structures are compatible with with laser desorption ionisation (LDI). The doped nano-particles adhere to constituents from the mark material and provide significant signal enhancement in SALDI-TOF mass spectrometry for compounds such as pharmaceuticals, nicotine, drugs of abuse, explosives and endogenous metabolites, when compared to corresponding signals from latent marks. Here, we present SALDI-TOF mass spectrometry intensity maps of endogenous phospholipid constituents within fingerprints following dusting with hydrophobic silica nano particles containing carbon black dopant. These compounds are being investigated as novel putative biomarkers to ascertain additional information that can identify the donor. A typical intensity map of a region of a fingerprint shows colours corresponding to the intensity of a single molecular mass for the compound under investigation. The points of highest intensities are represented as red spots. The high intensity spots are located along the ridge lines while the troughs between the ridges are shown in blue. Images appear to show that the source of the metabolites is the sweat pores, running along the ridges from which the compounds are secreted.

A02602-04441

Nanocomposite Engineering of Nanocrystalline MaterialsJackie Y. YING*Institute of Bioengineering and Nanotechnology, Singapore*

Nanocrystalline materials are of interest for a variety of applications. This talk describes the design and functionalization of nanocomposite materials for biological and chemical applications. Specifically, we have generated metallic, metal oxide and semiconducting nanocrystals for bioimaging, biolabeling, bioseparation, biosensing and catalysis. These nanocrystals are ≤ 10 nm in size, and are surface modified to provide for high dispersion, biocompatibility, and water solubility. They are used as building block to create multifunctional nanocomposite particles.

A02620-04480

Controllable Magnetic Bead Motion on Patterned NiFe Elliptical Lines for Biosensing ApplicationsCheolGi KIM; Anandakumar SARELLA;
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We have designed, fabricated and demonstrated a novel system for controllable translational magnetic bead motion on Si surfaces. The soft NiFe ellipses of size ($9 \mu\text{m} \times 4 \mu\text{m} \times 0.1 \mu\text{m}$) are lithographically patterned to transport magnetic beads. By the application of an external rotating magnetic field, NiFe ellipses can generate local inhomogeneous stray magnetic fields due to arrangement of the ellipses. Since the partially magnetized magnetic beads are attracted to poles of the saturated ellipses, the inhomogeneity in the stray magnetic fields on the patterned NiFe elliptical lines can direct the motion of the magnetic beads by rotating external magnetic field either in clockwise or counter clockwise directions. The magnetic beads can jump from one ellipse to another with respect to the external rotating magnetic field. We demonstrated the back and forth motion of the Dynabead® M-280 magnetic bead on the pathway. The back and forth motion of the magnetic beads on the patterned lines can be controlled by adjusting the elemental distance between the ellipses. The patterned soft NiFe thin film elements can be easily magnetized by the application of relatively weak magnetic field due to the low coercivity, thereby serving as a convenient template for flexible manipulation of magnetic beads. This technique enables micro-translocation of the magnetic beads coated with biomolecules to the specific binding sites of the surface and as well a drive off the non-specific binding molecules from the surface in performing number of sequential bead detection experiments for future integrated lab-on-a chip systems.

A02674-04592

The Sonochemical Synthesis of Functional NanomaterialsMuthupandian ASHOKKUMAR*School of Chemistry, University of Melbourne, Melbourne/Victoria, Australia*

The synthesis of functional nanomaterials is one of the active research areas as these materials are found useful in a variety of potential applications, such as, imaging, energy production, therapeutic and diagnostic medicine, etc. A number of advanced synthetic techniques have been developed for synthesizing nanosized metal and polymer particles. Sonochemistry is found to be a useful technique for synthesizing a variety of nanomaterials possessing specific physical and functional properties.

Sonochemistry refers to the chemical reactions that could be induced by the interaction of sound waves and gas bubbles in liquids. The interaction between (ultra)sound waves and dissolved gas nuclei results in a phenomenon known as acoustic cavitation, which involves the growth of existing gas nuclei by rectified diffusion and inertial collapse of resonance-sized microbubbles. The near adiabatic collapse generates very high temperatures and pressures within the cavitation bubbles leading to the formation of highly reactive radicals. In addition to generating these primary radicals, acoustic cavitation is also accompanied by a number of physical effects, such as agitation, microstreaming, enhanced mass transport, etc. The reactions between the primary radicals and solutes adsorbed at the bubble/solution interface may generate secondary radicals. The primary and secondary radicals and the physical effects generated during acoustic cavitation have been found useful in the synthesis of a variety of nanomaterials.

We have developed a sonochemical methodology for controlling the size and size distribution of metal nanoparticles. It is also possible to synthesize core/shell or alloy bimetallic nanoparticles that could be used in fuel cells and other applications. Recently, we have developed a simple and efficient technique for the encapsulation of magnetite nanoparticles within host latex particles. This ultrasound initiated procedure represents an effective means of producing a range of nanocomposites consisting of multiple combinations of different polymers and encapsulation materials. These nanocomposites can, in principle, be used in various biomedical, cosmetics and food processing applications for the controlled release of pharmaceuticals and flavours. An overview of the sonochemical synthesis of functional nanomaterials will be presented.

A02684-04616

Label-free Electrical Detection of DNA using Gold Nanoparticle Enhancement in Carbon Nanotube Networked Field-Effect Transistors

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The development of label-free and sequence-selective detection of deoxyribonucleic acid (DNA) based on electrochemical or electroluminescence methods and direct electrical detection using field-effect transistors (FETs) has become a subject of intense research. Carbon nanotubes (CNTs) have attracted much attention in this endeavor

because their conductance is sensitive to the molecular adsorption on tube walls or within tubes due to their unique electronic structures. In addition, CNTs are biocompatible and chemically stable in ambient environment. Therefore, there is a growing body of research on the development of CNT based devices for DNA sensing.

Typically, SWNT networked FETs (SNFETs) are able to detect ~ 1nM of DNA. In this communication, we report that the detection sensitivity of DNA for SNFETs can be further improved to ~ 100fM by using a “nanoparticle-enhancement” approach, where the target DNAs are hybridized with the probe DNAs on devices and the reporter DNAs labeled with Au-nanoparticles (AuNPs) flank a segment of the target DNA sequence. The mechanism studies indicated that the AuNP is attached with large number of reporter DNAs which provide very high density of negative charges to the junction and therefore alter the conductivity of SNFETs. This method provides a new approach for the enhancement of DNA selectivity in carbon nanotube networked field-effect transistors and we believe the sensitivity can be further improved by properly adjusting the composition of the SWNT networks.

A02773-04738

Electroconductive Hydrogels: Co-networks of Poly(HEMA-co-PEGMA-co-HMMA-co-MPC) and Polypyrrole (PPy) for Implantable Biosensors and Deep Brain Stimulation

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The long term *in-vivo* biocompatibility and electrical performance of implantable biosensors for physiological status monitoring and of indwelling electrodes for deep brain stimulation are of paramount importance in trauma care and as neuronal prostheses, respectively. Bioactive and electrically responsive hydrogels have emerged as a paradigm shift in the development of multifunctional, biorecognition membrane layers for these biomedical devices. Both polymeric constituents of the co-network may be molecularly engineered to confer predictable hydration levels, mechanical properties, surface chemistries, interfacial impedance properties and micro-nano-topologies that render them similar to and thus mimics of the tissue bed within which they are to be deployed. The recognition membrane layer of implantable biosensors requires that biorecognition molecules such as oxidoreductase enzymes, their co-factors, redox mediators and biomimetic moieties be purposefully incorporated at bio-functionally relevant levels within the hydrogel. We

have developed approaches to the design of these tissue-to-electrode interfaces based on the use of *biomimetic* electroconductive hydrogel *co-networks*. Hydrogels are rendered electroconductive through the *in-situ* synthesis of *electrically conductive* polymers or the incorporation of functionalized single walled carbon nanotubes within cross-linked, highly hydrophilic polymer networks. Our approach to biocompatibility employs *biomimicry* - the creation of *biocompatible* interfaces that emulate the chemistry and topography of the extracellular matrix and the modulus of the tissue bed within which the indwelling device must reside.

Hydrogel biointerfaces were formed from tetraethyleneglycol diacrylate (TEGDA) crosslinked hydroxyethylmethacrylate (HEMA). Polyethylene glycol methacrylate (PEGMA; 0.3 – 0.5 mol%) and 2-methacryloyloxyethyl phosphorylcholine (MPC; 5 – 10 mol%) were introduced to provide *in vivo* biocompatibility through their mitigation of adsorptive protein denaturation. These poly(HEMA-co-PEGMA-co-MPC) hydrogels exhibited an increase in the percent hydration with an increase in the MPC content (93.8% hydration at 10 mol% MPC). MPC is shown to have a larger hydrating influence when compared to PEG. FITC-dye tagged fibronectin (0.1 and 1.0 ng/ml at 25 °C) adsorbed via the Langmuir isotherm where K_d and Q_m , quantitatively confirmed the reduction in protein adsorption with hydration and with an increase in MPC content. Human aortic muscle endothelial cells had greater than 80% viability while human muscle fibroblasts showed high viability (> 80%) and low proliferation (< 40%). Hydrogels containing *N*-[tris(hydroxymethyl)methyl]-acrylamide (HMMA; 5 mol%) [p(HEMA)-co-PEGMA-co-HMMA hydrogels] that were cross linked with 1, 3, 5, 7, 9 and 12 M % TEGDA were found to hydrate in accordance with Flory-Huggins-Rehner theory. The release of the mediator, ferrocene monocarboxylic acid (FcCOOH), was via diffusion with diffusivities that ranged between 2.64×10^{-8} cm²/s (1 M% TEGDA) to 4.87×10^{-9} cm²/s (12 M% TEGDA) and showed thermally activated transport with activation energies that were 30 kJ/mol (3 M%) to 57 kJ/mol (12 M%). Polypyrrole (PPy) co-networks within these hydrogels provided interference screening of endogenous interferents in biosensor applications and resulted in biointerfaces with greatly reduced interfacial impedances.

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Selective Sensing of Biomaterials Using Metal-clad Waveguide Biosensor

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A number of techniques are being employed for the detection of micro-organisms including optical and

chemical. Among optical techniques, “waveguide (WG) biosensing” is appreciable due to its simplicity and enhanced sensitivity. We report the fabrication of a four layer metal clad waveguide (MCWG) biosensor known as dip type waveguide biosensor following the plasmon resonance which appears in the reflectance spectra. These biosensors improve the sensing characteristics of an adlayer thickness above one 150-200 nm. We employed Ag film or bi-metal film layers Au/Ag as cladding layer by physical vapour deposition at or below 10^{-6} torr. A 300 – 350 nm polystyrene or polyLlysine film was spin coated, which act as core medium. The change in the refractive index of cover medium (top cladding layer) changes the critical angle for total internal refraction at the resonance so causing changes due to different cover media. By monitoring changes in reflectance characteristics of the device, bio-molecules or biomaterials present in the cover medium (*E. coli* bacteria, antibody-antigen interaction *etc.*) can easily be sensed. We demonstrate that bi-metal clad waveguide show enhanced sensitivity compared with single metal clad WG. The sensitivity is even enhanced if the thicknesses of two metals are comparable. This is due to presence of two Plasmon modes in two metal layers, which can resonate to produce enhanced fields. The work has been extended to immobilization of antibodies (IgG) as or on the top cladding layer is on under way. These have been utilized to introduce specificity into MCWG and show promising results. In this paper, we will also discuss the response of WG biosensor to different antibodies.

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A General Approach to Generate Multifunctional Nano-architectures from DNA-based ABC Monomers for Biosensing and Drug-delivery-coupled Imaging

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Assembling different moieties onto a single core building block can create synergistic functions and enable real world applications including intelligent sensing and imaging. However, precise attachment of different moieties requires that the core block is both multivalent and anisotropic. In this talk, I will focus on our creation of Anisotropic, Branched, and Crosslinkable building blocks (ABC monomers) from which multifunctional nano-architectures have been assembled. A target-driven polymerization process has also been developed based on ABC monomers. In this novel process, multiple signals within each ABC monomer get amplified upon polymerization, and polymers are generated only in the presence of a specific DNA molecule, enabling highly-sensitive pathogen detection. Using this ABC monomer system, we have also designed a biocompatible multi-drug delivery vector that delivers both drugs and imaging-tracers simultaneously. Our

ABC monomer approach provides a general yet versatile route towards creating a range of multifunctional nano-architectures that can be used in multiplexed biosensing and drug-delivery-coupled imaging.

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Simulation of the Thickness Dependency of Enzyme Mediator Variations within Hydrogel Biorecognition Membranes

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In vivo biotransducers, when implanted in the body, generally face the inflammatory foreign body response and eventual fibrous encapsulation. The use of polymeric hydrogel materials containing specific bioactive moieties such as polyethylene glycol (PEG) and phosphorylcholine (PC) are being developed to mitigate this problem. The hydrogels also function as membrane reservoirs for immobilized enzymes in enzymatic biosensors. Such membranes therefore combine molecular recognition (enzymatic) and bioactive biocompatibility (PEG and PC) in a responsive device. We investigate, by electrochemical finite element modeling, the performance of highly hydrated bioactive hydrogels containing immobilized GOx and/or LOx enzymes and immobilized ferrocene monocarboxylic acid (FcCOOH) mediator as the biorecognition layer in voltammetric and amperometric biosensors.

A dual sensing electrochemical cell on a chip has been microfabricated on a glass substrate using gold as the electrode metal and silicon nitride as the passivation layer. The manufactured chip is then functionalized to facilitate covalent linkage between hydrogel and the chip surface. The biorecognition layer was fabricated from an ca. 1.0-5.0 microns thick, spun-applied, UV-polymerized, interpenetrating network of principally tetraethylene glycol (TEGDA) cross-linked poly(2-hydroxyethyl methacrylate) (HEMA) that contained a methacrylate-derivatized pyrrole component, 2-methacryloyloxyethyl-4(3-pyrrolyl)butanate (MPB), and two biomimetic methacrylate components; one with poly(ethyleneglycol) (110) (PEG) chains and the other with phosphorylcholine (PC) moieties, both pendant to the polymer backbone. The mediator was physically entrapped or covalently immobilized via covalent tethering using UV-polymerizable monomers, Fc-AEMA and Fc-PEG(3500)-AEMA, to produce pendant redox moieties within the hydrogel.

The amount of oxidoreductase enzyme and redox mediator loaded within the bioactive hydrogel membrane and cast onto the chip surface influences the stability and morphology of the hydrogel layer and has a direct bearing on

the sensitivity of the resulting biotransducer. Higher levels of enzyme loading were seen to yield surface discoloration and unstable hydrogel layers. In this simulation study, we seek the optimal loading of the enzyme and mediator concentration within the hydrogel. The finite element modeling was performed using COMSOL Multiphysics Modeling software (v.3.5a). In this model a thin layer of hydrogel membrane of defined thickness and containing enzyme and mediator of defined concentrations and diffusion coefficients was considered as sitting on top of a single microdisc electrode. The geometry was then solved with the electrode reaction kinetics obeying Butler-Volmer kinetics and the diffusion of species obeying Fick's law. The enzyme was assumed to obey Michaelis-Menten kinetics. Preliminary studies indicate good agreement between the model results and theoretical expectations. Further investigation is underway to optimize the effect of enzyme and mediator loading, enzyme/mediator ratio and hydrogel thickness at a particular loading level.

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