Celebrating 25 Years of IMRE: Research Highlights on Nanomaterials and Nanotechnologies

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The Institute of Materials Research and Engineering (IMRE) is a research institute of the Science and Engineering Research Council (SERC), Agency for Science, Technology and Research (A*STAR). IMRE was established in September 1997. Over the past 25 years, IMRE has developed core competence and interdisciplinary teams for material development from fundamental discoveries to industrial translation. Currently, with over 400 researchers and state-of-the-art research facilities, IMRE conducts world class research in important material and material technology fields, including polymer composites, optical materials, electronic materials, soft materials, structural materials, energy materials, biomaterials, quantum technologies, as well as advanced characterization.

As a material-centered research institute in Singapore, IMRE has been playing important roles in pushing science boundaries and developing cutting-edge technologies. One of the key strategies is to partner international organisations, research institutes and industry to fulfil its vision to be a leading research institute to accelerate materials research, moving from "Made in Singapore" towards "Created in Singapore".

Nanotechnology is one of the key enabling technologies of the 21st century. It has important role to play in international effort in sustainability.[1] IMRE's nanomaterials R&D encompasses a wide range of activities, including nanomaterial synthesis (0D, 1D, 2D, and 3D), nanocomposite formulation, nanostructures/nanodevices fabrication, modeling, and characterization. The fields of application include optics/photonics, electronics/optoelectronics, medicine, computing, energy, agriculture, aquaculture, and food. To show case the recent advances in nanomaterial and nanotechnology research in IMRE and with their collaborators, we are pleased to bring you this special virtual issue in *ACS Nano* commemorating the 25th anniversary of the institute (**Figure 1**). This special virtual issue captures the selected research work published in ACS Nano in the last five to ten years by leading experts in IMRE and their collaborators, in the fields of (1) Nano-Optics and Nano-Photonics, (2) Nanobiotechnologies, (3) Two Dimensional Materials and Devices, (4) Two Dimensional Materials for Energy and Catalysis, and (5) Advanced Nano Characterization.

Figure 1. Celebrating 25 years of innovate.

NANO-OPTICS AND NANO-PHOTONICS

Nano-optics or nano-photonics is a research field that investigates the interaction between light and nanostructures for various applications, such as optical holography, high resolution color printing, beam shaping, functional optoelectronic devices, etc. IMRE and collaborators have made significant contributions in the development of nanostructures with optical resonances, including metallic nanoantennas with localized plasmon resonance,[2,-7] dielectric nanoantennas with Mie resonances,[8-11] hybrid plasmon-Mie resonance,[12] and leaky waveguide mode.[13-16] Zhaogang Dong and Joel K. W. Yang et al., designed Sb_2S_3-based antenna arrays, where the corresponding designed Mie resonances in the visible regime could be tuned in a reversible manner (**Figure 2A**).[11] These tunable Sb_2S_3 nanoantenna arrays could enable the next generation of high-resolution reflective color displays, hologram displays, and compact LiDAR systems. Vasilii Mylnikov, Arseniy I. Kuznetsov, and co-workers designed a single subwavelength GaAs nanocylinder with the supercavity modes to achieve nano lasers, where the nanocylinder has a diameter of 500 nm and a height of 330 nm (**Figure 2B**). [10]

A hybrid metal-dielectric nanoantenna has the potential to realize the advantages of the Purcell enhancement effect in metallic nanostructures, while achieving the strong scattering directivity and low Ohmic losses of dielectric nanostructures. Jinfa Ho, Arseniy I. Kuznetsov, Joel K. W. Yang, and co-workers designed a compact hybrid nanoantenna with the Yagi-Uda configuration (**Figure 2C**).[12] The hybrid antenna exhibits a high directivity with an ultra-small footprint of only 0.38 λ^2. Such nanoantenna has good potential for applications, such as on-chip optical communications. In addition, camouflage is critical for many animals to survive in the natural world. Kun Huang, Zhaogang Dong, and co-workers demonstrate an optical masquerade, as inspired by masquerade in nature, with perturbative silicon metasurfaces that could camouflage a designed object (**Figure 2D**).[8] The presented design of optical masquerade is potentially applicable for optical security, anti-counterfeiting, and encoding.

In plasmonic resonators studies, the team reported a theoretical model based on an annular ring-like light source to simulate the dark-field illumination conditions accurately.[2] They fabricated metallic gold nanostructures with the lateral gap plasmons for enhancing the second-harmonic generation (SHG) emission. [3] They established the correlation between the structural density and absorption cross-section in a plasmonic system, where it is able to achieve saturated colors by fabricating aluminum plasmonic nanostructures in a metal–insulator–metal configuration with the gap plasmons.[17] Most recently, Hong Liu and his team devised a 3D woodpile photonic crystal to support high-order optical modes with the side illumination and it allows one-step printing of 3D photonic structural colors without requiring post-processing or subwavelength features (**Figure 2E**).[16]

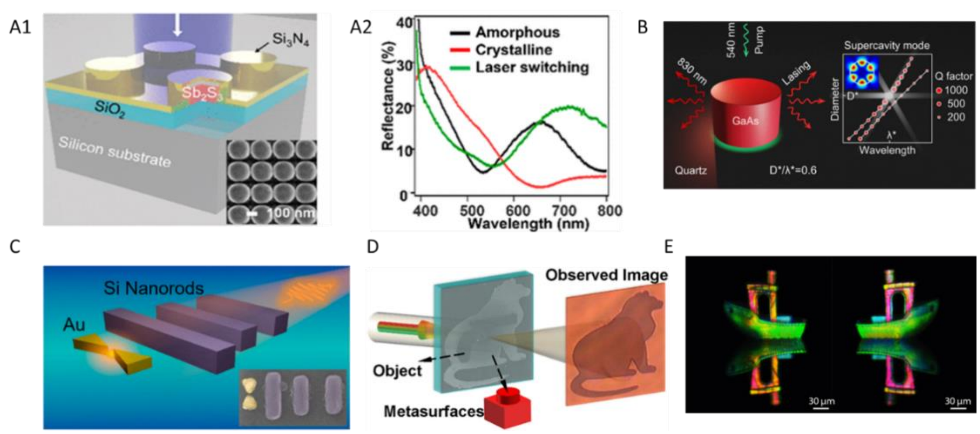

Figure 2. (A) Reversible tuning of Mie resonances in the visible spectrum. Reprinted with permission from ref 11. Copyright 2021 American Chemical Society. (B) Lasing action in single subwavelength particles supporting Supercavity Modes. Reprinted with permission from ref 10. Copyright 2020 American Chemical Society. (C) Highly directive hybrid metal–dielectric Yagi-Uda nanoantennas. Reprinted with permission from ref 12. Copyright 2018 American Chemical Society. (D) Bio-inspired photonic masquerade with perturbative metasurfaces. Reprinted with permission from ref 8. Copyright 2020 American Chemical Society. (E) High-order photonic cavity modes enabled 3D structural color. Reprinted with permission from ref 16. Copyright 2022 American Chemical Society.

NANOBIOTECHNOLOGIES

Nanomaterials' distinct optical and physical properties have been largely exploited for medical and healthcare applications. IMRE has made significant contribution in exploiting nanomaterials for sensing, [18, 19] diagnostic, [20-22] imaging, [23-26] and therapy.[27] Research activities are revolved around the synthesis of nanoparticles and biofunctionalization of the nanoparticles for desired applications. Tedrick Thomas Salim Lew, Xiaodi Su, and Lisa Ng, etc have recently demonstrated B-cell immunogenic epitope-functionalized gold nanoparticles as serological sensing element for SARS-CoV-2 IgG detection (**Figure 3A**).[20] This is a one-step, single reagent colorimetric COVID-19 IgG test that provides rapid quantitative detection (<30 min turnover time) with much improved selectivity against IgG of other coronavirus, e.g. SARS, MERS, and HCoV-HKU1. Heng Li Chee, David Paramelle and co-workers reported biocompatible peptide-coated ultrasmall superparamagnetic Iron Oxide Nanoparticles (USPION) for *in vivo* contrast-enhanced Magnetic Resonance Imaging (MRI).[23] From a library of 21 peptides and a total of 86 different peptide-coated USPIONs, they identified a candidate that is highly stable against electrolyte-induced aggregation with less nonspecific binding to cells and no cellular toxicity. This nanoparticle reagent is benchmarked to commercially available MRI contrast agents for its superior performance. Another work related to nanoparticle for *in vivo* application was reported by Bin Liu, Ben Zhong Tang, and Deling Kong, etc.[26] They reported organic nanodots with aggregation-induced emission characteristics (AIE dots) for long-term tracking of adipose-derived stem cells (ADSCs) (**Figure 3B**). The AIE dots possess high fluorescence (QY 25±1%), excellent biological and photophysical stabilities, and low *in vivo* toxicity, superior to the most popular commercial cell trackers.

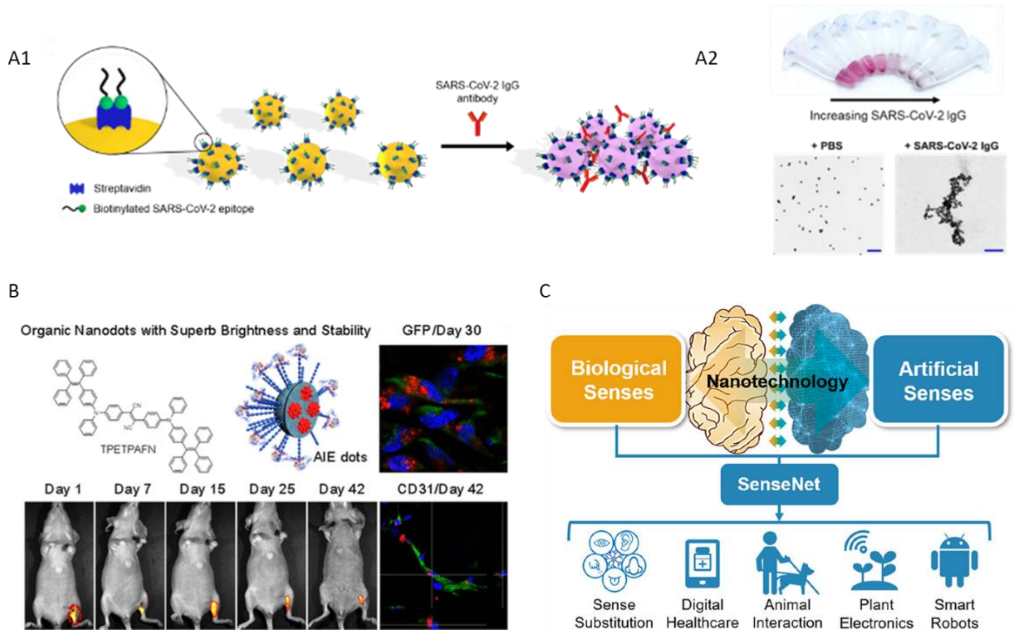

Figure 3. (A) B-cell immunogenic epitope-functionalized gold nanoparticles for rapid and selective detection of SARS-CoV-2 IgG Antibodies. Reprinted with permission from ref 20. Copyright 2021 American Chemical Society. (B) Superb bright and stable organic nanodots for precise and long-term tracking of adipose-derived stem cells. Reprinted with permission from ref 26. Copyright 2014 American Chemical Society. (C) A review on artificial sense technology. Reprinted with permission from ref 18. Copyright 2021 American Chemical Society.

In additional to the exploitation of functional colloidal nanoparticles, IMRE also possess the state-of-art nanofabrication infrastructures for making biologically patterned surfaces and devices.[28-33] Karen Chong, Graham Leggett, and co-workers developed micrometer scale protein patterning technologies by interferometric lithography (IL) and nanoimprint lithography (NIL).[28] This technique is highly beneficial for fabricating biological nanostructures without involving sophisticated nanofabrication infrastructures. In addition, nanomaterials' potential for emulating the biological senses of human being (touch, feel, smell, healing, etc) has attracted extensive attention, Xiaodong Chen, Xian Jun Loh, and co-workers have recently outlined the advantages of artificial sense technology and the potential exploitation of nanomaterials (**Figure 3C**).[18]

TWO DIMENSIONAL MATERIALS AND DEVICES

Atomically-thin two-dimensional (2D) materials, such as graphene, boron nitride (BN), and transition metal dichalcogenides (TMDCs, e.g., MoS_2, WS_2 etc), represent an emergent and divergent class of nanostructured low-dimensional materials. The distinct properties of 2D materials, originating from the reduction in the dimensionality, makes them well-suited to applications where bulk materials would otherwise be unsuitable. [34-36] In recent years, IMRE and our collaborators have made a number of important scientific findings and technology developments in the field of 2D materials research. These research include material growth/synthesis, non-destructive 2D doping, hybrids formation, electrical and optoelectric property characterization, 2D device fabrication, etc. [37-48] Yee-Fun Lim, Swee Liang Wong, Dongzhi Chi and co-workers reported a reactive-barrier-based approach for the growth of monolayer molybdenum disulfide (MoS_2) on sapphire, which are highly crystalline and well-oriented, leading to the observation of well-resolved electronic band structure and spin-orbit splitting even at room temperature (**Figure 4A**).[43] Using photoemission spectroscopy, Fabio Bussolotti, Kuan Eng Johnson Goh, and co-workers revealed a clear reduction in MoS_2 core level and valence band binding energies that is dependent on the density of S-vacancies within a MoS_2 monolayer interfaced with graphite (**Figure 4B**).[44] Their results highlight the importance of vacancy density and electrostatic disorder in TMDC-based electronics for improving device performance and production. On the other hand, the major limitation of unipolar conduction in n-type TMDCs was overcome by Weifeng Yang, Hao Gong, Shijie Wang, and Dongzhi Chi *et al*. where highly reactive nitrogen atoms was demonstrated to achieve effective substitutional *p*-type doping in mono- and few-layer WS_2 without introducing detrimental lattice distortion owing to its low kinetic energy (**Figure 4C**).[45] Yu Li Huang, Andrew Thye Shen Wee, and co-workers fabricated an intrinsic leteral heterojunction within a monolayer (1L) tungsten diselenide (WSe_2), where the *p*-doping for WSe_2 originated from a molecular acceptor layer, providing insights into employing surface doping, for example, in designing lateral organic TMDC heterostructures for future devices **(Figure 4D)**. [46]

Another vital element for TMDCs devices is the metal-semiconductor interface in which undesirable contact resistances have hindered the progress of their electronic/optoelectronic and quantum applications. To this end, Chit Siong Aaron Lau and Kuan Eng Johnson Goh *et al.*, revealed that indium alloy contacts can significantly improve performance in chemical vapor deposition grown WS_2 devices owing to reduced contact resistances (**Figure 4E**).[47] The team further addressed the poor recombination efficiency of TMDs at higher excitation energies owing to band-nested suppression by designing a self-assembled lanthanide (Ln)/TMDC hybrid based on a cost-effective and efficient solution-processed approach (**Figure 4F**).[48] The emission energy from Ln^{3+} sources can be effectively transferred to the TMDC monolayers under low power exposure (0.13 mW) at room temperature, activating the characteristic monolayer fluorescence in place of Ln^{3+} emission signatures. Their study extends the functionalities of TMDCs for optoelectronic and photonic applications.

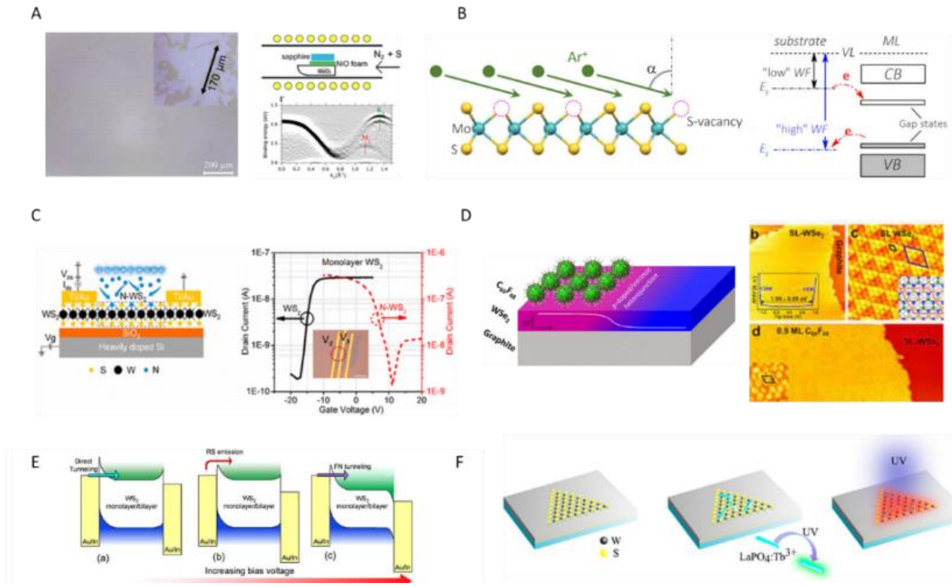

Figure 4. (A) Modification of vapor phase concentrations in MoS_2 growth using a NiO foam barrier. Reprinted with permission from ref 43. Copyright 2018 American Chemical Society. (B) Impact of S-vacancies on the charge injection barrier at the electrical contact with the MoS_2 monolayer. Reprinted with permission from ref 44. Copyright 2021 American Chemical Society. (C) Direct n- to p-type channel conversion in monolayer/few-layer WS_2 field-effect transistors by atomic nitrogen treatment. Reprinted with permission from ref 45. Copyright 2018 American Chemical Society. (D) Electronic properties of a 1D intrinsic/p-doped heterojunction in a 2D transition metal dichalcogenide semiconductor. Reprinted with permission from ref 46. Copyright 2017American Chemical Society. (E) Quantum transport in two-dimensional WS_2 with high-efficiency carrier injection through indium alloy contacts. Reprinted in part with permission from ref 47. Copyright 2020 American Chemical Society. (F) Band nesting bypass in WS_2 monolayers *via* Förster Resonance Energy Transfer. Reprinted with permission from ref 48. Copyright 2020 American Chemical Society.

TWO DIMENTIONAL MATERIALS FOR ENGINERGY AND CATALYSIS

In addition to the electronics and optoelectronics properties, the catalytic properties of 2D materials have also been extensively exploited in energy and catalysis research. [49-52] MXenes, a class of 2D inorganic compounds, including metal carbides, nitrides, and carbonitrides, has recently emerged as promising candidates for large-area catalytic energy storage and conversion. They have distinct hydrophilicity and, high metallic conductivity. They are also easy to produce by solution processing. Kang Rui Garrick Lim, Babak Anasori and Zhi Wei She etc reviewed recent strategies in the design of MXene hybrids for industrially relevant electrocatalytic, photocatalytic, and photoelectrocatalytic applications, such as water splitting, metal-air/sulfur batteries, carbon dioxide reduction, and nitrogen reduction (**Figure 5A**).[49] They discussed MXene hybrids for their significant enhancement of the catalytic performances beyond the sum of their individual components. They clarified the roles of individual material components in the MXene hybrids, and thereafter provided material design strategies for highly efficient and durable catalytic applications. In a research paper, the team presented a simple and scalable method to circumvent adventitious oxidation in Mo_2CT_x MXenes via *in-situ* sulfidation to form a Mo_2CT_x/2H-MoS_2 nanohybrid (**Figure 5B**).[50] This study is important for practical implementation of MXenes as noble metal free catalysts for applications in water splitting and energy conversion.

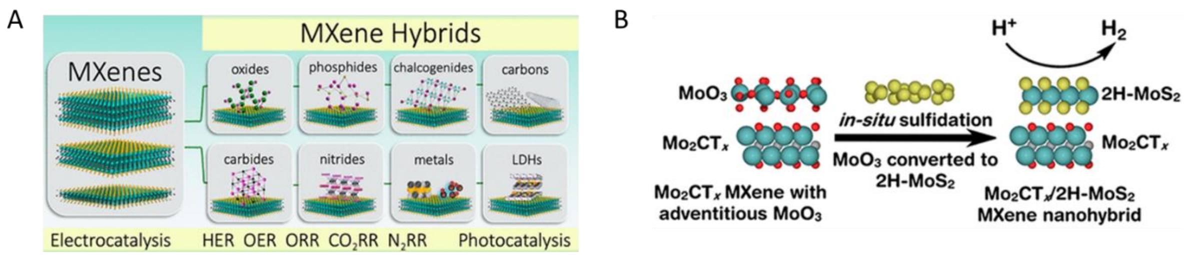

Figure 5. (A) A review on 2D MXene hybrids and nanocomposites for catalytic energy storage and conversion. Reprinted with permission from ref 49. Copyright 2020 American Chemical Society. (B) 2H-MoS_2 on Mo_2CT_x MXene nanohybrid for efficient and durable electrocatalytic hydrogen evolution. Reprinted with permission from ref 50. Copyright 2020 American Chemical Society.

ADVANCED NANO CHARACTERIZATION

IMRE is equipped with state-of-the-art characterization facilities and expertise. Our researchers develop revolutionary characterization capabilities through instrumentation, modeling, and combinational analysis involving whole spectrum of characterization and analytical tools. Examples include in-situ analysis of galvanic replacement reactions of silver nanocubes and Au studied by liquid-cell Transmission Electron Microscopy (TEM), [53] accurate modeling of dark-field scattering spectra of plasmonic nano structures [54], and high-resolution TEM analysis of atomically thick dielectric coatings. [55] IMRE's materials analysis and characterization capabilities and tools are openly accessible by A*STAR researchers and nation-wide research community, such as universities, public sector agencies and industry.

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Notes

The authors declare no competing financial interests.

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