

Division of Synthetic Chemistry

– Advanced Inorganic Synthesis –

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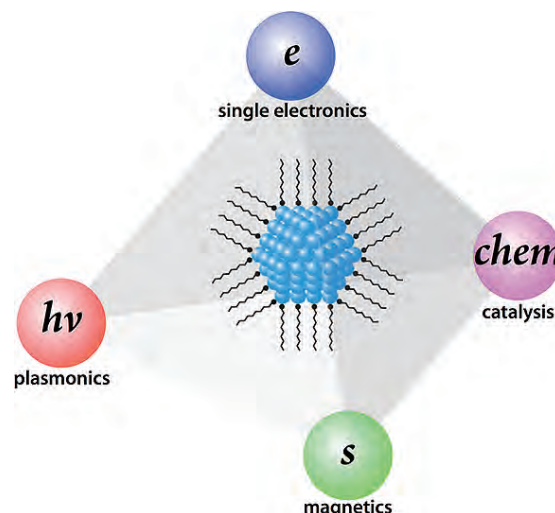
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Scope of Research

We are focusing on the precise synthesis of inorganic nanoparticles by controlling the primary (size, shape, composition, *etc.*) and secondary (spatial arrangement) structures to tune properties such as electron confinement, carrier oscillation, spin, and catalysis. These high-quality inorganic nanoparticles are applied to both high-performance nanodevices (*e.g.*, single electron transistor, plasmon waveguide, and nanocomposite magnet) and photo-energy conversion materials (*e.g.*, overall water splitting and solar cell).

KEYWORDS

Inorganic Nanoparticles Single Electronics Plasmonics
Nanocomposite Magnet Photocatalysts



Selected Publications

Saruyama, M.; Kim, S.; Nishino, T.; Sakamoto, M.; Haruta, M.; Kurata, H.; Akiyama, S.; Yamada, T.; Domen, K.; Teranishi, T., Phase-Segregated NiP_x@FeP_yO_z Core@Shell Nanoparticles: Ready-to-Use Nanocatalysts for Electro- and Photo-Catalytic Water Oxidation through *in-situ* Activation by Structural Transformation and Spontaneous Ligand Removal, *Chem. Sci.*, **9**, 4830-4836 (2018).

Lian, Z.; Sakamoto, M.; Matsunaga, H.; Vequizo, J. J. M.; Yamakata, A.; Haruta, M.; Kurata, H.; Teranishi, T., Near Infrared Light Induced Plasmonic Hot Hole Transfer at a Nano-Heterointerface, *Nat. Commun.*, **9**, 2314 (2018).

Eguchi, D.; Sakamoto, M.; Teranishi, T., Ligand Effect on the Catalytic Activity of Gold Clusters in the Electrochemical Hydrogen Evolution Reaction, *Chem. Sci.*, **9**, 261-265 (2018).

Number of Surface-attached Acceptors on a Quantum Dot Impacts Energy Transfer and Photon Upconversion Efficiencies

Photon upconversion, which converts low-energy photons into high-energy photons, is attractive for various applications including solar energy conversion, bioimaging, and optogenetics. Compared with upconversion using lanthanide-based nanocrystals, triplet–triplet annihilation upconversion (TTA-UC) can occur under a relatively low light flux comparable to that of sunlight. Quantum dots (QDs) are excellent light-harvesting materials that efficiently absorb light ranging from the ultraviolet to the infrared region. The quantum yield of triplet–triplet energy transfer (TTET) from QDs to suitable organic molecules has been demonstrated to reach almost 100%. Moreover, the energy loss induced by singlet–triplet energy gaps (ΔE_{ST}) can be decreased by the characteristic small singlet–triplet splitting ($\Delta E_{ST} < 15$ mV) of QDs. Therefore, composites of QDs and organic molecules have emerged as an ideal TTA-UC system that can potentially reach higher efficiency. However, the UC efficiency of QD-molecular composites is still lower than that reported for a molecular system. The tailored approach promoting the UC efficiency using QDs has been required. Herein, we discovered that the number of acceptor molecules (n) on a single QD dictates the efficiencies of triplet–triplet energy transfer (TTET) and TTA-UC. Systematic investigation of different types of QDs (CdSe, CdTe) coordinated with precisely determined numbers of perylene-3-carboxylic acid (Pe) ligands revealed that both TTET and TTA-UC efficiencies are determined by the number of Pe on a QD. Based on our results, we demonstrated that the intrinsic TTET efficiency (Φ_{TTET}/n) independent of the number of acceptor molecules is a good evaluation guide of TTA-UC systems. The proper choice of QDs and acceptors providing both sufficient free energy change for TTET and high n are important to achieve efficient TTA-UC.

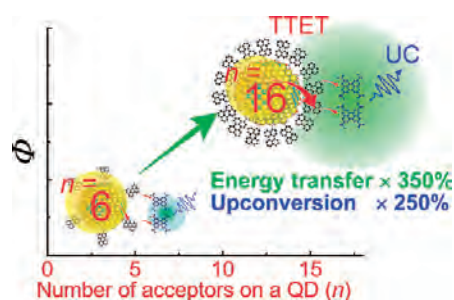


Figure 1. Illustration of triplet–triplet annihilation upconversion (TTA-UC) using QD-molecular composites. The number of molecules on a QD determines the efficiencies of triplet–triplet energy transfer (TTET) and TTA-UC. (Adapted with permission from ACS Photonics 2020, 7, 1876-1884. Copyright 2020 American Chemical Society.)

Self-activated Rh–Zr Mixed Oxide as a Nonhazardous Cocatalyst for Photocatalytic Hydrogen Evolution

Photocatalytic water splitting is a desirable technology for sustainable hydrogen production. Various kinds of photocatalysts have been developed for effective utilization of solar energy. In addition, developing cocatalysts for photocatalysis is also indispensable for improving photocatalytic activity; this is because the cocatalysts can promote extraction of excited charges from the photocatalysts and reduce the activation energy of the water splitting reaction. Though RhCrO_x is commonly used because of their excellent activity, long-time photo-irradiation causes elution of Cr(VI) ions, resulting in not only decreased activity but also environmental damage and health hazards. Here we demonstrate that a Rh–Zr mixed oxide works as an efficient cocatalyst for hydrogen evolution. Impregnation of Zr and Rh complexes (Zr/Rh = 5 wt/wt%) formed RhZrO_x mixed oxide cocatalyst particles on Al-doped SrTiO_3 , which showed 31 times higher photocatalytic water splitting activity than a RhO_x cocatalyst. X-ray photoelectron spectroscopy revealed that the dissociation of Cl^- ions from preformed Rh–Cl–Zr–O solid led to formation of the active phase of RhZrO_x . Electrochemical measurements showed that addition of Zr suppresses not only the ORR, but also the HER of Rh. This fact determines the proper weight ratio of Zr/Rh (= 5) in RhZrO_x for water reduction reaction. Additional CoO_x loading as a water oxidation cocatalyst further improved the activity by 120%, resulting in an apparent quantum yield of ca. 33% at 365 nm and a good durability in at least 60 h. This novel nonhazardous Zr-based cocatalyst will be useful for large-scale solar-hydrogen production.

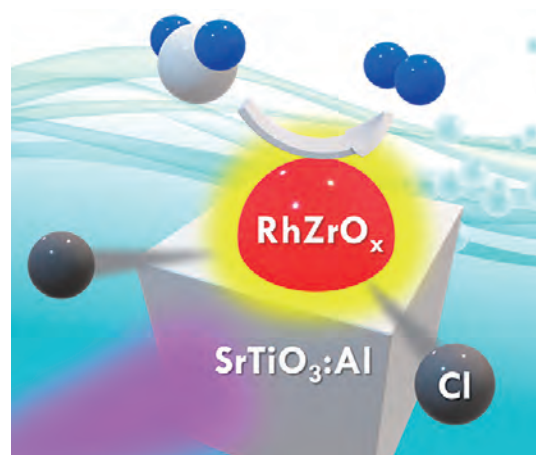


Figure 2. Illustration of RhZrO_x cocatalyst on $\text{SrTiO}_3:\text{Al}$ photocatalyst. RhZrO_x works as an active site for photocatalytic hydrogen evolution reaction. Active RhZrO_x cocatalyst is formed by the spontaneous Cl^- removal from preformed Rh–Cl–Zr–O solid during photocatalysis.