HIGHLIGHTS FROM RECENT LITERATURE

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Each issue of *Gold Bulletin* contains key highlights from the research and patent literature. Authors who publish highquality work in other journals are invited to send a copy of their publication to the editor for inclusion in the next issue.

Nanotechnology

Multiple strategies to activate gold nanoparticles as antibiotics

Widespread antibiotic resistance calls for new strategies. Nanotechnology provides a chance to overcome antibiotic resistance by multiple modes of antibiotic mechanisms. This feature article from leading Chinese researchers (Zhao and Jiang, Nanoscale, 2013, Accepted Manuscript doi:10.1039/C3NR01990J) reviews the progress of activating gold nanoparticles with non-antibiotic or antibiotic molecules to combat bacterial resistance, analyzes the gap between experimental achievements and the real clinical application, and suggests some potential directions in developing antibacterial nanodrugs.

Patterned transparent conductive Au films through direct reduction of gold thiocyanate

Construction of structurally defined patterned metal films is a fundamental objective in the emerging and active field of bottom-up nanotechnology. A new strategy for constructing macroscopically organized Au nanostructured films has been presented by Israeli researchers (Advanced Functional Materials, Early View, doi:10.1002/adfm.201300881). The approach is based upon a novel phenomenon in which incubation of water-soluble Au(SCN)₄^{1–} complex with amine-displaying surfaces gives rise to spontaneous crystallization and concurrent reduction, resulting in the formation of

patterned metallic gold films. The Au films exhibit unique nanoribbon morphology, likely corresponding to aurophilic interactions between the complex moieties anchored to the amine groups through electrostatic attraction. Critically, no external reducing agents are needed to initiate or promote formation of the metallic Au films. In essence, the thiocyanate ligands provide the means for surface targeting of the complex, guide the Au crystallization process, and, importantly, donate the reducing electrons. It is shown that the Au films exhibit electrical conductivity and high transparency over a wide spectral range, lending the new approach to possible applications in optoelectronics, catalysis, and sensing. In a broader context, a new gold chemistry route is presented in which ligand-enabled crystallization/reduction could open the way to a wealth of innovative reaction pathways and applications.

Complete light annihilation in an ultrathin layer of gold nanoparticles

Swedish researchers have demonstrated that an incident light beam can be completely annihilated in a single layer of randomly distributed and widely spaced gold nanoparticle antennas. Under certain conditions, each antenna dissipates more than ten times the number of photons that enter its geometric cross-sectional area. The underlying physics can be understood in terms of a critical coupling to localized plasmons in the nanoparticles or, equivalently, in terms of destructive optical Fano interference and so-called coherent absorption. See Svendahl et al., Nano Lett., Article ASAP doi:10.1021/nl400849f

A novel dengue virus detection method that couples DNAzyme and gold nanoparticle approaches

Recent epidemics of dengue viruses (DENV) coupled with new outbreaks on the horizon have renewed the demand for

novel detection methods that have the ability to identify this viral pathogen prior to the manifestation of symptoms. The ability to detect DENV in a timely manner is essential for rapid recovery from disease symptoms. A modified lab-derived 10-23 DNAzyme tethered to gold nanoparticles provides a powerful tool for the detection of viruses, such as DENV. University of Notre Dame researchers examined the effectiveness of coupling DNAzyme (DDZ) activation to the salt-induced aggregation of gold nanoparticles (AuNP) to detect DENV progeny in mosquito cells. A DNAzyme was designed to recognize the 5' cyclization sequence that is conserved among all DENV, and conjugated to AuNPs. DDZ-AuNP has demonstrated the ability to detect the genomic RNA of our model dengue strain, DENV-2 NGC, isolated from infected Aedes albopictus C6/36 cells. These targeting events lead to the rapid aggregation of AuNPs, resulting in a red to clear color transition of the reaction mixes, and thus positive detection of the DENV RNA genome. The inclusion of SDS in the reaction mixture permitted the detection of DENV directly from cell culture supernatants without additional sample processing. Specificity assays demonstrated detection is DENV specific, while sensitivity assays confirm detection at levels of 1× 101 TCID50 units. These results demonstrate DDZ-AuNP effectively detects DENV genomes in a sequence-specific manner and at concentrations that are practical for field use. The team has developed an effective detection assay using DNAzyme catalysis coupled with AuNP aggregation for the detection of DENV genomes in a sequencespecific manner. Full development of our novel DDZ-AuNP detection method will provide a practical, rapid, and low-cost alternative for the detection of DENV in mosquito cells and tissues, and possibly infected patient serum, in a matter of minutes with little to no specialized training required. See Virology Journal 2013, 10:201 doi:10. 1186/1743-422X-10-201

Two-color laser printing of individual gold nanorods

German researchers report on the deposition of individual gold nanorods from an optical trap using two different laser wavelengths (*Nano Lett.*, Article ASAP. doi:10.1021/nl401788w). Laser light, not being resonant to the plasmon resonances of the nanorods, is used for stable trapping and in situ alignment of individual nanorods. Laser light, being resonant to the transversal mode of the nanorods, is used for depositing nanorods at desired locations. The power and polarization dependence of the process is investigated and discussed in terms of force balances between gradient and scattering forces, plasmonic heating, and rotational diffusion of the nanorods. This two-color approach enables faster printing than its one-color equivalent and provides control over the angular orientation ($\pm 16^{\circ}$) and location of the deposited nanorods at the single-nanorod level.

Polyelectrolyte-coated gold magnetic nanoparticles for immunoassay development: toward point of care diagnostics for syphilis screening

Immediate response for disease control relies on simple, inexpensive, and sensitive diagnostic tests, highly sought after for timely and accurate test of various diseases, including infectious diseases. Composite Fe₃O₄/Au nanoparticles have attracted considerable interest in diagnostic applications due to their unique physical and chemical properties. Here, Chinese researchers introduce a simple coating procedure for gold magnetic nanoparticles (GMNs) with poly(acrylic acid) (PAA) (Anal. Chem., 2013, 85 (14), pp 6688-6695 doi:10. 1021/ac400517e. PAA-coated GMNs (PGMNs) were stable and monodispersed and characterized by Fourier transforminfrared spectroscopy, transmission electron microscopy, UVvisible scanning spectrophotometry, thermogravimetric analysis, and Zetasizer methodologies. For diagnostic application, they established a novel lateral flow immunoassay (LFIA) strip test system where recombinant Treponema pallidum antigens were conjugated with PGMNs to construct a particle probe for detection of anti-Tp antibodies. Intriguingly, the particle probes specifically identified Tp antibodies with a detection limitation as low as 1 national clinical unit/mL. An ample pool of 1,020 sera samples from three independent hospitals were obtained to assess our PGMNs-based LFIA strips, which exhibited substantially high values of sensitivity and specificity for all clinical tests (higher than 97 %) and, therefore, proved to be a suitable approach for syphilis screening at a point-of-care test manner.

Gold coating for a high-performance $Li_4Ti_5O_{12}$ nanorod aggregates anode in lithium-ion batteries

An effective strategy by combination of alcoholysis, solid-state reaction, and coating techniques has been reported to prepare Au@Li₄Ti₅O₁₂ nanorod aggregates as anode materials for Liion batteries (Wang et al., Journal of Power Sources, 10.1016/j. jpowsour.2013.06.156). The lithium diffusion coefficient of resulting Au@Li₄Ti₅O₁₂ is 7.32×10^{-10} cm² s⁻¹, and its stable reversible capacity is 169 mAh g⁻¹ with the retention of 91.1 % after 100 cycles at 5 °C. Moreover, it also exhibits excellent ratecapability performance. The superior cycling performance can be attributed to the unique nanorod characteristics, structural stability, and the improved ionic and electronic conduction in the electrode due to the uniform nano-coating of Au.

Nanometer-scale thermometry in a living cell

Sensitive probing of temperature variations on nanometer scales is an outstanding challenge in many areas of modern science and technology. In particular, a thermometer capable of sub-degree temperature resolution over a large range of temperatures as well as integration within a living system could provide a powerful new tool in many areas of biological, physical, and chemical research. Possibilities range from the temperature-induced control of gene expression and tumor metabolism to the cell-selective treatment of disease and the study of heat dissipation in integrated circuits. By combining local light-induced heat sources with sensitive nanoscale thermometry, it may also be possible to engineer biological processes at the subcellular level. Here, US and Chinese researchers demonstrate a new approach to nanoscale thermometry that uses coherent manipulation of the electronic spin associated with nitrogen-vacancy color centers in diamond (Kucsko et al., Nature 500, 54-58 (2013) 10.1038/nature12373). This technique makes it possible to detect temperature variations as small as 1.8 mK (a sensitivity of 9 mK $Hz^{-1/2}$) in an ultrapure bulk diamond sample. Using nitrogen-vacancy centers in diamond nanocrystals (nanodiamonds), the local thermal environment was measured on length scales as short as 200 nm. Finally, by introducing both nanodiamonds and gold nanoparticles into a single human embryonic fibroblast, temperature-gradient control and mapping at the subcellular level was demonstrated, enabling unique potential applications in life sciences.

Electronics

Thin sputter deposited gold films on In_2O_3 :Sn, SnO₂:In, TiO₂, and glass: Optical, electrical, and structural effects

Thin gold films are promising transparent conductors with many actual and potential uses in "green" technologies, transparent electronics, etc. These applications require different substrate materials, and hence it is important to understand the role of the substrate on Au thin film growth. Such effects have been studied in this work by a Swedish team, wherein Au films-ranging from island structures, via large-scale coalescence into meandering metal networks, to thin homogenous layers-were deposited by DC magnetron sputtering onto glass substrates and In₂O₃:Sn (ITO), SnO₂:In and TiO₂ base layers backed by glass (Lansåker et al., Solar Energy Materials and Solar Cells, 10.1016/j.solmat.2013.06.051 doi:10.1016/ j.solmat.2013.06.051#doilink). Optical, electrical, and structural properties were recorded for films deposited onto unheated substrates. Distinct and characteristic differences in Au growth on the various backings were found. Thus, ITO and SnO₂:In base layers yielded gold films with island features remaining to larger thicknesses than for deposition directly onto glass, and the sheet resistance was lower for gold deposition onto SnO₂:In and ITO only when the gold films were less than ~5 nm in thickness. These results highlight the complexity of substrates' influence on thin film formation.

High-performance solution-processed organic transistors with electroless-plated electrodes

Electroless-plated gold and platinum films are used as source and drain electrodes in high-performance solution-processed organic field-effect transistors (OFETs), representing a promising large-area, near room temperature, and vacuum-free technique to form low-resistance metal-to-semiconductor interfaces in ambient atmosphere. Developing nondisplacement conditions using a Pt-colloidal catalyst for soft electroless plating, the electrodes are deposited on crystallized thin films of 2,9-didecyl-dinaphtho[2,3-b:2',3'-f]thieno[3,2-b]thiophene (C10-DNTT) without significant damage to the semiconductor material, as described by Japanese scientists (Ito et al., Organic Electronics, 10.1016/j.orgel.2013.05.006 doi:10.1016/j. orgel.2013.05.006#doilink). The top-contact OFETs show remarkable performance, with a mobility of 6.0 cm² V⁻¹ s⁻¹. The method represents a practical fabrication technique to mass-produce circuitry arrays of nearly best-performing OFETs for the printed electronics industry.

Gold-platinum-palladium alloy bonding wire

Tanaka researchers describe a gold-platinum-palladium alloy bonding wire for vehicle-mounted semiconductors, which exhibits excellent reliability in the connection with aluminum pads, while being highly reliable when left at high temperatures, even in cases where an epoxy resin containing no halogen substance is used, thereby capable of maintaining the electrical characteristics after being left at high temperatures (patent number WO2013080851). This wire contains 0.4–1.2 % by mass of platinum, 0.01–0.5 % by mass of palladium, 10–30 ppm by mass of aluminum, and 10–60 ppm by mass of calcium and/or magnesium in total, with the balance made up of gold having a purity of 99.999 % by mass or more.

Fuel cells

Microbial biofuel cell operating effectively through carbon nanotube blended with gold—titania nanocomposites modified electrode

In this study, Chinese researchers have explored the possibility to fabricate microbial biofuel cell operating with carbon nanotube–gold–titania nanocomposites (CNT/Au/TiO₂) as anode modifier (Wu et al., Electrochimica Acta, 10.1016/j. electacta.2013.07.166 doi:10.1016/j.electacta.2013.07.166# doilink). The results demonstrate that the CNT/Au/TiO₂ electrode could be utilized as a new and effective microbial fuel cell (MFC) anode, which integrate the advantages of relevant nanocomposites such as high conductivity, high-specific surface area, and easy adsorption of the microorganism. It is evident that the three-dimensional network nanostructures of CNT/Au/TiO₂ are propitious to improve the relevant anode surface area and, thus, the adsorption of the microorganism, which can efficiently promote the electronic transfer rate between the probe and electrode. Meanwhile, it is noted that open circuit voltage of the CNT/Au/TiO₂ nanocomposites modified carbon paper anode increased to 0.77 V, which is more than twice that of the open circuit voltage obtained with bare carbon paper anode (0.36 V). And the MFC equipped with CNT/Au/TiO2 nanocomposites modified carbon paper anode delivers a maximum power density of 2.4 mW m⁻², which is three times larger than that obtained from the MFC with bare carbon paper. This observation illustrates that the CNT/Au/TiO₂ nanocomposites modified electrode could obviously increase the relevant electron transfer rate and promotes the electron exchange at electrode surface, which could readily provide enhanced stability and relatively long life-span to facilitate the high electricity production efficiency, suggesting its promising prospect application in MFCs.

Fuel cell separator and method for producing same

Japanese researchers describe a fuel cell separator having a region comprising a stainless steel substrate, a gold-plated layer that is formed on the stainless steel substrate, and has pinholes and a stainless steel passivation layer formed inside the pinholes, wherein the gold-plated layer and the stainless steel substrate come into contact without involving the stainless steel passivation layer (patent number CN103069629). This fuel cell separator can be produced by forming a strike gold-plated layer on the surface of the stainless steel substrate by using an acidic gold strike plating solution, then forming the gold-plated layer, and subsequently performing passivation treatment. The present invention provides a fuel cell separator that has excellent corrosion resistance and that can be manufactured inexpensively, and a method for producing the fuel cell separator.

Catalysis

Bimetallic Au–Cu supported on ceria for PROX reaction : effects of Cu/Au atomic ratios and thermal pretreatments

The influence of pretreatment (calcination or reduction) on bimetallic Au–Cu/CeO₂ catalysts with a fixed Au loading of 2 wt.% and an atomic Cu/Au ratio of 1/3, 1/1, or 3/1 was studied by researcher groups based in France and China (Applied Catalysis B: Environmental. In Press, 10.1016/j. apcatb.2013.05.010). The catalysts were prepared by direct anionic exchange of HAuCl₄ followed by a Cu(NO₃)₂ impregnation. Both pretreatment methods led to catalysts of significantly different structures and surface compositions. For the calcined samples, the presence of a copper oxide phase can be observed when the Cu/Au is 3/1, whereas for the reduced catalysts, there is the formation of an Au-Cu alloy and of metallic copper due to the immediate decomposition of the copper precursor, which is able to be partially alloyed with gold. XPS reveals that the calcination led to the segregation of a CuO-rich phase on the surface, while in the case of reduction, the surface is not so Cu-enriched. All of the catalysts were evaluated by preferential oxidation of CO in the presence of hydrogen and compared with monometallic 2 % Au/CeO2 and 2 % Cu/CeO2. For all catalysts, it was found that irrespective of the atomic Cu/Au ratio, a reduction improves the CO conversion and CO₂ yield compared with monometallics, while enhanced catalytic performances are only observed for the 1/3 Cu/Au atomic ratio of the calcined samples. The reduced Au-Cu bimetallics are stable with time on-stream contrary to the calcined one where the formation of carbonate-like species, responsible of a deactivation, is observed. Furthermore, the reduced catalyst remained active even in the presence of CO₂ and H₂O in the gas stream

Photocatalytic water splitting on Au/TiO₂ nanocomposites synthesized through various routes: enhancement in photocatalytic activity due to SPR effect

Au/TiO₂ nanocomposites were prepared by in situ, photodeposition (PD), and SMAD method. The synthesized photocatalysts were evaluated for their photocatalytic activity in hydrogen generation. The hydrogen evolution rate was observed to be 1,200, 920, and 1,600 μ mol h⁻¹ for in situ, photodeposition (PD), and SMAD, respectively, under UV-Vis light illumination. However, under purely visible light illumination, the highest hydrogen evolution of 32.4 µmol h⁻¹ was observed in case of Au/TiO₂ prepared by PD followed by SMAD (6.9 μ mol h⁻¹). The significantly high photocatalytic activity as demonstrated by Au/TiO₂ synthesized by photodeposition method under purely visible light irradiation proves the potential of photocatalyst in efficient solar energy conversion. The polydisperse particles of AuNPs on TiO₂ by photodeposition method enhance plasmonic effects. The catalyst was thoroughly characterized by powdered X-ray diffraction, UV-Vis DRS, and TEM for understanding the plasmonic properties. TEM images further substantiated the deposition of gold on TiO₂ matrix of heterogeneous particle size with an average size of 8-10 (smaller particle) and 53-70 nm (bigger particles). The photodeposition of AuNPs on different titania supports was also studied by the American and Indian researchers (Applied Catalysis B: Environmental. In Press, 10. 1016/j.apcatb.2013.05.057 doi:10.1016/j.apcatb.2013.05.057# doilink).

Diesel oxidation catalyst

This patent, lodged by Umicore scientists (patent number US2013202509), relates to a catalytically active material

consisting of an inner core (1) and an outer shell (2) surrounding this core; the core being formed from palladium and gold fixed together on a first support oxide, and the shell comprising platinum fixed on a second support oxide to a diesel oxidation catalyst comprising this catalytically active material, and to an exhaust gas cleaning system comprising this diesel oxidation catalyst.

Oxidation of cycloalkanes in the presence of a supported bimetallic gold-palladium catalyst

This patent (patent number WO2013114330) relates to a process for the oxidation of cycloalkanes utilizing a supported gold and palladium catalyst and the use of the supported gold and palladium catalyst for the oxidation of cycloalkanes. Also described is a process for the preparation of the supported catalyst.

Preparation of copper oxide-cerium oxide-supported nano-gold catalysts and its application in removal of carbon monoxide in hydrogen stream

A preparation method of nano-gold catalysts supported on copper oxide-cerium oxide (CuO-CeO2) and a process of preferential oxidation of carbon monoxide by oxygen in hydrogen stream with the nano-gold catalysts have been disclosed by Taiwanese researchers (US2013183221). CuO-CeO2 is prepared by either coprecipitation or incipientwetness impregnation method, and gold is deposited thereon by deposition-precipitation. After adding CuO into Au/CeO2, the interaction between the nano-gold and the support is increased, thereby enhancing the stability of the gold particle and the activity of the catalysts. Preferential oxidation of CO in hydrogen stream (with O2 existing) over these catalysts is carried out in a fixed bed reactor. The O2/CO ratio should be between 0.5 and 4. The catalyst is applied to remove CO (to lower than 10 ppm) in hydrogen stream in fuel cell to prevent from poisoning of the electrode of the fuel cell.

Nano-gold catalysts on Fe-modified ceria for pure hydrogen production via WGS and PROX: effect of preparation method and Fe-doping on the structural and catalytic properties

Two series of Fe-doped (5, 10, and 20 wt.% Fe₂O₃) ceria oxides were synthesized by mechanochemical mixing (MM) and impregnation (IM) methods, as reported by European researchers (Ilieva et al., Applied Catalysis A: General. 10. 1016/j.apcata.2013.07.014 doi:10.1016/j.apcata.2013.07. 014#doilink. The Au catalysts (3 wt.%) supported on these mixed oxides were tested in WGSR and PROX. Gold catalysts on supports prepared by IM exhibited WGS activity lower than that of gold on undoped ceria. Significantly better WGS

performance was demonstrated over gold catalysts on supports synthesized by MM. The observed differences in the catalytic behavior were explained by the crucial role of the gold dispersion depending on the preparation method. The highest WGS activity of Au5FeCeMM catalyst was attributed to the cooperative catalytic work of Fe-modified ceria and magnetite with the highest extent of non-stoichiometricity. In contrast to WGSR, the catalytic behavior in PROX over gold catalysts on supports prepared by both methods did not differ significantly. Fe likely acted as a ceria modifier because no changes in Fe-phases and Fe-valence state were observed upon reaction. The nanosized hematite particles covering the ceria grains in the case of the IM method could diminish the ceria basicity explaining the observed good and stable PROX performance over Au10FeCeIM catalyst in the presence of CO₂ and water.

Exceptional oxidation activity with size-controlled supported gold clusters of low atomicity

The catalytic activity of gold depends on particle size, with the reactivity increasing as the particle diameter decreases. However, investigations into behavior in the sub-nanometer regime (where gold exists as small clusters of a few atoms) began only recently with advances in synthesis and characterization techniques. Here, Spanish and US-based researchers report an easy method to prepare isolated gold atoms supported on functionalized carbon nanotubes and their performance in the oxidation of thiophenol with O₂ (Corma et al., Nature Chemistry (2013). 10.1038/nchem.172). It is shown that single gold atoms are not active, but they aggregate under reaction conditions into gold clusters of low atomicity that exhibit a catalytic activity comparable to that of sulfhydryl oxidase enzymes. When clusters grow into larger nanoparticles, catalyst activity drops to zero. Theoretical calculations show that gold clusters are able to activate thiophenol and O₂ simultaneously, and larger nanoparticles are passivated by strongly adsorbed thiolates. The combination of both reactants activation and facile product desorption makes gold clusters excellent catalysts.

Chemistry

Gold(I) complexes of tetrathiaheterohelicene phosphanes

New tetrathia[7]helicene-based gold(I) complexes have been readily prepared by reaction of phosphine ligands with Au(tht)Cl at two molar ratios. These complexes have been fully characterized by analytical and spectroscopic techniques by the German and Italian researchers (Cauterrucio et al., *Inorg. Chem.*, 2013, *52* (14), pp 7995–8004, doi:10.1021/ic4005533), as well as quantum chemical calculations. The molecular structure of one of the dinuclear complexes has

been determined by single-crystal X-ray diffraction, showing a gold–gold interaction of 3.1825(3)Å and a significant contraction of the 7-TH total dihedral angle. This complex displays luminescence emission at room and low temperature in diluted solution and in the solid state. Quantum chemical calculations show that the luminescence emission at room temperature is primarily due to slightly perturbed fluorescence emission from purely $\pi\pi$ * excited states of the conjugated helicene scaffold. At 77 K, phosphorescence emission is displayed as well. Preliminary studies on the use of two of these complexes as catalysts in typical Au(I)-catalyzed cycloisomerizations have demonstrated the reactivity of these systems in the intramolecular allene hydroarylations and the hydroxycarboxylation of allene-carboxylates.

Luminescent gold(III) compounds for organic lightemitting devices and their preparation

The described invention provides a class of luminescent gold(III) compounds containing a tridentate ligand with one strong σ -donating group. Also provided are methods for synthesizing these compounds, as well as uses of these compounds as electrophosphorescent materials in phosphorescent organic light-emitting devices to provide electroluminescence (patent number US20130193428).

Synthesis of new gold(I) thiolates containing amino acid moieties with potential biological interest

Spanish researchers describe the reaction of a gold(I) complex [Au(SpyCOOH)(PPh₃)], which contains nicotinic acid thiolate, with several amino acid esters such as glycine methyl ester or the enantiomerically pure L-isomers of alanine methyl ester,

phenylalanine methyl ester, valine methyl ester, methionine methyl ester, and proline methyl ester produces the gold(I) derivatives with the new thiolate containing amino acid ester ligands [Au{SpyCONHCH(R)COOMe}(PPh₃)] (Gutierrez et al., Inorg. Chem., 2013, 52 (11), pp 6473-6480 doi:10. 1021/ic4003803). The reaction of these amino acid ester derivatives with LiOH in methanol and acidification with KHSO₄ until pH 3-4 afford the corresponding acids, which are water-soluble species. These amino acid compounds can be further coupled with other amines, such as, for example, isopropylamine, to give the corresponding amide derivatives. The species with glycine methyl ester and valine methyl ester have been characterized by X-ray crystallography, showing, in the second case, only one of the enantiomers, which proves that retention of the configuration after reaction occurs.

Hydrophenoxylation of alkynes by cooperative gold catalysis

According to UK-based researchers, the title method gives aryl vinyl ethers in high yields, short reaction times, Z-stereospecificity, and good regioselectivities. Insights into the reaction mechanism highlight the role of $[{Au(NHC)}_2 (\mu$ -OH)][BF₄] (NHC = N-heterocyclic carbene), as both a Lewis acid [Au(NHC)][BF₄] and a Brønsted base [Au(NHC)(OH)], thereby generating a synergistic effect between the two gold moieties (Oonishi et al., Angewandte Chemie International Edition, doi:10.1002/anie.201304182).

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