Part 1: Critical Reviews

Introduction of the year 2021

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In the present chapter we introduce the volume and its three sections, namely critical reviews, highlights on recent "hot" topics and SPR lectures in photochemistry. The most significant editorial releases in photochemistry in 2021 are resumed therein by presenting awards, special issues, reviews and handbooks.

1 Introduction

The present volume marks a symbolic but extremely significant milestone in the Photochemistry series of the Specialist Periodical Reports published by the Royal Society of Chemistry, which dates back to the year 1970 when it was conceived from an idea of D. Bryce-Smith (see Fig. 1a).[1] This periodical has now reached the half-century milestone, thanks to the efforts of motivated photochemists, who steered the wheel of this editorial endeavour with the invaluable support of the Royal Society of Chemistry. The list of these scientists includes, among others, professors A. Gilbert[2] and A. Albini[3] (a complete list of the editors of the SPR is provided in Table 1).

Volume 50 follows the subdivision in three different sections which was recently introduced in the series. The first part of this book includes a series of critical reviews on the recent advances in computational (D. Roca Sanjuan, University of Valencia) and organic photochemistry, focused on the reactivity of different functional groups (J. Perez Prieto, University of Valencia and S. Crespi, Uppsala University), of alkenes (K. Mizuno, Osaka Prefecture University) and aromatic compounds (T. Tsuno, Nihon University), as well as recent discoveries in transition metal photocatalysis (C. Lambruschini, University of Genova). The second part of the volume includes highlight papers on recent "hot" topics, in a format aimed to provide the readership with a selection of the advanced research that could be also a pleasant reading for practitioners. In this framework, the present issue collects reviews on visible light driven-enantioselective processes (Prof. J. Aleman), the formation of C–calchogen bonds (Profs D. Qiu and X. Xia, Peking University), the photochemical activation of chloroarenes (Dr. M. Giedyk, Polish Academy of Sciences), the synthetic application of photoelectrocatalysis (Profs D. Ravelli, University of Pavia and L. Dell'Amico, University of Padova), an overview on photovoltaic techniques (Dr. J. Milic, Adolphe Merkle Institute) and on the computational analyses of fluorescence absorption spectra (Dr. D. Loco, Qubit Pharmaceuticals) Finally, the "SPR Lectures in

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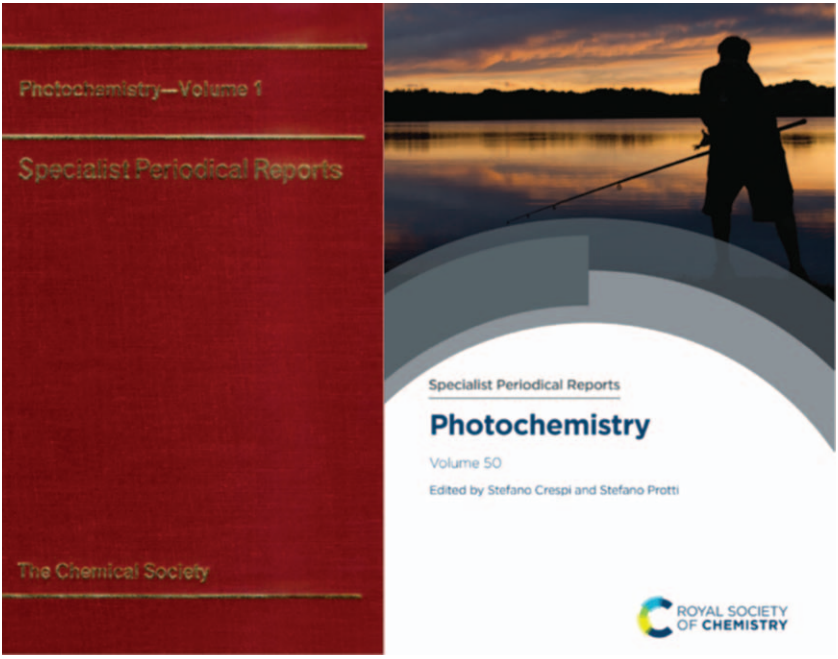


Fig. 1 (a) Front Matter of the Specialist Periodical Reports in Photochemistry, vol. 1 (edited by Derek Bryce-Smith) and (b) vol. 50. Reproduced with permission from the Royal Society of Chemistry, Copyright 1970 and 2022.

Table 1 Editors of the specialist periodical reports in photochemistry since its debut (1970).

Derek Bryce-Smith, University of Reading (1970–1994)
Andrew Gilbert, University of Reading (1989–2002)
Ian Dunkin, University of Strathclyde, Glasgow (2003–2007)
Angelo Albini, University of Pavia (2009–2019)
Elisa Fasani, University of Pavia (2012–2017)
Stefano Protti, University of Pavia (2017–current)
Carlotta Raviola, University of Pavia (2020)
Stefano Crespi, Uppsala University (2021–current)

Photochemistry'' of this year have been authored by Profs B. Feringa (University of Groningen), N. Zaccheroni (University of Bologna), T. Banerjee (Pilani Campus, Rajasthan) and T. Yoon (Madison University). This format allows the more established researchers in the field to provide an introduction and a more scholastic point of view on the concepts of the chosen topic, aiming at an academic and student readership in photochemistry.

2 Awards and prizes

Five years after the Nobel Prize in Chemistry received by B. Feringa, F. Stoddard and J.-P. Sauvage another photochemist, Prof. David MacMillan (Princeton University), was honoured by this appointment, for the fundamental contribution to organocatalysis.[4–7] In December 8, 2021 the

Scottish scientist pronounced the Nobel Lecture entitled ''Asymmetric organocatalysis: Democratizing catalysis for a sustainable world''.[8]

Prof. Paolo Melchiorre, (ICIQ, Tarragona) was awarded with the 2021 Pedler Award of the RSC Organic Division in recognition of his efforts in the development of asymmetric photocatalytic protocols.[9] The 2021 Corday–Morgan Prizes went to Prof. Junwang Tang, of University College London, in view of the discovery of efficient photocatalytic materials to be used in solar-to-fuel synthesis[10,11] and to Prof. Jan Verlet, (Durham University), for the excellent results obtained in the development of novel innovative methods to probe the fundamental physical chemistry underpinning electron-molecule reactions.[12,13]

The PhotoCube™ reactor from ThalesNano, which is the result of a partnership between ComInnex, ThalesNano, and Professor Timothy Noël (Technical University of Eindhoven, now University of Amsterdam) was the winner of the R&D Top 100 competition, as the first batch and flow-based benchtop multi-wavelength reactor commercially available for advanced photochemical processes.[14]

Prof. Laura Gagliardi (University of Chicago) was awarded with the Faraday Lectureship Prize for the contributions to the development of multireference quantum chemical strategies to investigate catalytic reaction and processes *via* excited state.[15,16]

Dr Pola Goldberg Oppenheimer (University of Birmingham) received the Beilby Medal, for the impressive studies on unconventional lithographic structuring of applied materials and the use optical spectroscopy in diagnosis.[17,18] The 2021 winner of the De Gennes Prize (RSC Material Chemistry Division) was Prof. Chad Mirkin (Northwestern University) for the invention and development of methods for nanolithography[19] characterized by high-area rapid printing, and for his contribution in supramolecular chemistry, and in particular for photocontrol in nanoparticle synthesis.[20] Prof. Jean-Luc Brédas (University of Arizona) received the 2021 Centenary Prize Winner for the impressive efforts in the investigation of the electronic properties of organic materials for electronics and photonics.[21,22]

The *Sir Geoffrey Wilkinson Award* was assigned to Dr James Wilton-Ely from the Imperial College London for the successful design of selective fluorescent probes for CO sensing in air and monitoring of this gas in cells.[23]

3 Handbooks

The handbook ''*Prebiotic Photochemistry: From Urey–Miller-like Experiments to Recent Findings*'' (Editors: F. Saija and G. Cassone) aims to provide the reader with a description of the state-of-the-art of such research field, as well of a series of analysis on the synergic contribution of light and other energy sources or selective conditions/media.[24]

The preparation of graphitic carbon nitride (gCN) materials and their applications in photocatalysis for solar-to-fuel conversion and environmental remediation is considered a highly promising area. The nine chapters volume ''Carbon Nitride Nanostructures for Sustainable Energy

Production and Environmental Remediation'' is the first handbook devoted on this topic and analyses in detail the different gCN nanostructures reported in literature and their versatile uses.[25] The volume 47 of the series ''Advances in Photosynthesis and Respiration'' (J.-R. Shen, K. Satoh and S. I. Allakhverdiev Editors, Springer) is focused on all aspects of natural and artificial photosynthesis, ranging from the structure of light-harvesting complexes and the role of far red chlorophylls.[26] On the other hand, the book ''Photosynthesis, Respiration, and Climate Change'' (Springer, 389 pp., also a part of the ''Advances in Photosynthesis and Respiration'' collection, edited by K. M. Becklin, J. K. Ward and D. A. Way adopted a more bio-oriented approach, and aims to integrate physiological and ecological perspectives on plant physiology with chapters on the recent advances in bioengineered photosynthesis.[27] I. Mohd, I. Ahamed and E. Lichtfouse are the editors of ''*Water Pollution and Remediation: Photocatalysis*'', whose chapters describes in detail the advanced photocatalytic techniques applied for the removal of pollutants, by providing a series of critical reviews on the advantages (and disadvantages) of photocatalysis for wastewater treatment, purification, and desalination.[28]

''Nanomaterials and Photocatalysis in Chemistry: Mechanistic and Experimental Approaches'' is the title of the book authored by M. Bilal, T. Khalid and N. Riaz, a discussion on solar driven heterogeneous photocatalysis that include several examples on the degradation of organic pollutants such as pharmaceuticals, additives and dyes.[29] Springer recently published also the multichapter ''*Handbook of Nanomaterials and Nanocomposites for Energy and Environmental Applications*'' (O. Vasilievna, K. Leticia, M. Torres-Martínez and B. I. Kharisov eds, 3786 pp., Springer), a comprehensive guide to the fields where such materials have been successfully employed, including (photo)catalysis, metallurgy, coatings, energy storage and environmental remediation.[30] In the aim of demonstrating the progresses in the structural, electronic and electrical properties of photocatalytic materials that can be achieved *via* introduction of defects, V. Gurylev wrote ''*Nanostructured Photocatalyst via Defect Engineering*'', an investigation on the nature of intrinsic and extrinsic defects in semiconductor photocatalytic materials.[31] A. M. Alvertis with ''*On Exciton–Vibration and Exciton–Photon Interactions in Organic Semiconductors*'' aimed to describe the physical mechanisms that allow for the generation and transfer of energy at the nanoscale in semiconductors, with the formation of excitons, that act as energy carriers.[32] The massive volume ''*Handbook of Laser Micro- and Nano-Engineering*'', edited by a team composed by K. Sugioka, G. Bo, M. Brandt, Y. Cheng, K. Du, C. Guo, M. Hong, L. Li, Y. Lu, A. Ostendorf, L. V. Zhigilei and M. Zhong, is a detailed description of the recent advances in ultrafast laser processing, with different examples of practical and commercial applications.[33]

The main spectroscopy and diffraction techniques currently applied to cultural heritage research have been reviewed in ''*Spectroscopy, Diffraction and Tomography in Art and Heritage Science*'', with particular attention to mobile equipment, large-scale instruments and infrastructural methods.[34]

Finally, we would like to cite an instruction manual recently published by the Royal Society of Chemistry on the enemies of the light *par excellence.* "*Vampirology: The Science of Horror's Most Famous Fiend*" by K. Harkup started from the stories found in most cultures across the globe and investigated how a scientific interpretation can shed light on the phenomena of the vampire myth.[35]

4 Special issues, collections and reviews

A Joint Special Virtual Issue entitled "*Photons at Play: Photocatalysis in Sustainable Chemistry*" was assembled by *ACS Sustainable Chemistry & Engineering* and *ACS Catalysis*, with the contribution of S. Protti (University of Pavia), T. Yoon (Madison University) and H. Han (Dalian Institute of Chemical Physics), a selection of papers published by the two journals in the 2018–2020 period.[36] Among the different contributions, C. B. Park reported the use of Amorphous Carbon Nitride as a visible light photocatalyst for the regeneration of regeneration of nicotinamide cofactor (NADH) in the biocatalytic conversion of α-ketoglutarate to L-glutamate.[37]

With the editing work of Dirk Guldi (Friedrich-Alexander-Universität Erlangen-Nürnberg) and Rik Tykwinski (University of Alberta), *ChemPhotoChem* recently dedicated a volume on singlet fission,[38] namely the interaction of a molecule in its singlet excited state with another one of the same species but in its ground state to generate two triplets, that was recently proposed as a strategy to overcome the upper thermodynamic limit for single-junction solar cells. Great attention has been given to computational experiments, that have been exploited by E. Pradhan *et al.* for designing singlet fission chromophores based on the diazadiborinine framework.[39]

ChemPhotoChem also launched the virtual collection "*Photocatalytic and Photoelectrochemical Carbon Dioxide Reduction*" edited by H. Huang (China University of Geosciences), G. Li (University of New Hampshire) and M. Schwalbe (Humboldt Universität zu Berlin), that focused on the recent development of light driven strategies in the conversion of carbon dioxide to chemical fuels.[40] Among the different contributions, Bellardita *et al.* reviewed and compared the electrocatalytic and photocatalytic methods described to achieve this target, and highlighted the synergism observed after the coupling of the two technologies.[41] In another paper, P. Gotico *et al.* investigated, by means of time resolved spectroscopy analyses, the potential of the Triazole-Linked Ruthenium–Rhenium Dyad Ru–cT–Re (Fig. 2) in the one-photon visible light reduction of CO_2.[42]

V. Augugliaro, V. Loddo, M. Bellardita, G. Marcì (University of Palermo) and S. Yurdakal (Afyon Kocatepe University) are the guest editors of the "*Photo(thermo)catalysis*" thematic issue published by Catalysis Today in honour of Prof. L. Palmisano[43] that has been co-author of more than 300 scientific papers, five handbooks, two didactical books and seven patents. Within the 268 pages of the issue, Dell'Edera *et al.* presented a review on the applications of photocatalytic nanostructured TiO_2 based coating, including microbial inactivation and air remediation.[44]

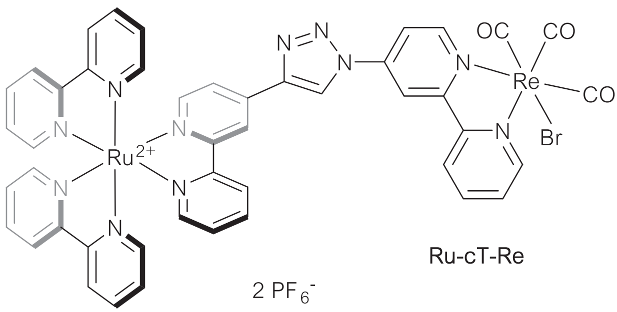
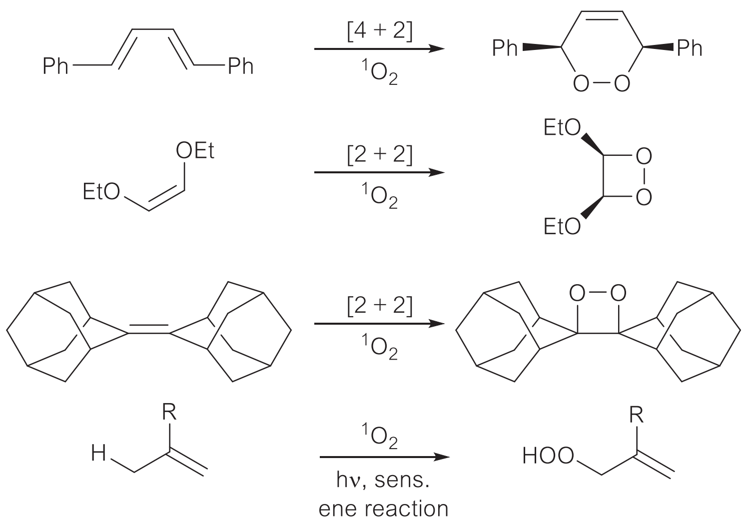


Fig. 2 Triazole-linked ruthenium–rhenium dyad Ru–cT–Re.[42]



Scheme 1

As the senior editor of *Photochemical and Photobiology*, Alexander Greer (Brooklyn College, NY) edited the special issue dedicated to Prof. Edward L. Clennan for his retirement.[45] Most contributions have been written by former group members, collaborators and colleagues of Prof. L. Clennan, pointing out the impressive contribution of this scientist in photocatalysis, singlet oxygen generation and photooxidation. In particular, Prof. M. Orfanopoulos reviewed the main classes of reaction of singlet oxygen with unsaturated molecules, namely the $[4+2]$ and the $[2+2]$ cycloaddition as well as the ene oxyfunctionalization of alkenes (resumed in Scheme 1). The mechanistic insight of processes has been examined at the light of the most recent experimental and computational data.[46]

M. Trotta (IPCF-CNR Istituto per i Processi Chimico Fisici, Bari) assembled a Photochemical and Photobiological collection on the application of hybrid and semiartificial photosynthetic systems,[47] including the ones involving photoanodes consisting of polyquinoid dyes onto mesoporous tin oxide surface[48] and photosynthetic reaction centre proteins covalently bounded to porous silicon pillars (PSiP).[49]

As the previous issue of 2016,[50] the volume 228 of the *Faraday Discussion* series is dedicated to "*Time-resolved Imaging of Photo-induced Dynamics*", and examines the new experimental techniques (including table-top based attosecond laser sources) that enable to monitor photo-induced processes with unprecedented temporal and spatial resolution. The contributions have been presented to the 2021 Faraday Discussions meeting that, due to the pandemic emergency, took place online.[51]

An issue of *Photochemistry and Photobiology* has been focused on the different aspects of germicidal photobiology and infection control, including the dosimetry of UV-C radiation exposure and carcinogenesis action spectra of germicidal ultraviolet radiation.[52] The preparation of the thematic volume has been preceded in 2020 by the virtual symposium "*New Developments in UV Germicidal Photobiology for Infection Control*" where R. S. Bergman presented an overview of the most used germicidal UV sources and apparatuses, and their potential application in the activation of viruses.[53] The unprecedented fast response by the global scientific community to the Covid-19 pandemic has inspired the topical collection entitled "*Tackling the global threats caused by pathogens: New horizons for antimicrobial Photodynamic Therapy*", that was assembled by K. Plaetzer (University of Salzburg), S. Nonell (University Ramon Llull, Barcelona), C. Viappiani (University of Parma) and T. Maisch (University Hospital Regensburg) and dedicated to Prof. Michael R. Hamblin on the occasion of his retirement.[54] Among the different contributions, we would like to point out a report on the different applications of photodynamic therapy in disinfection published by Aroso *et al.*[55] and a detailed investigation on a formulation containing Chlorin e6 and Sodium magnesium chlorophyllin as a selective photofungicide (Fig. 3).[56]

The 2021 edition of *Pharmaceuticals* dedicated to Photodynamic Therapy has been edited by Prof. S. Mordon (University of Bordeaux)[57] and counts 14 published papers, including a detailed investigation of the photophysics (lifetime emission, fluorescence quantum yield) of Protoporphyrin IX, Pyropheophorbide-a, and Photofrin photosensitizers (Fig. 4) in solvents of different nature.[58]

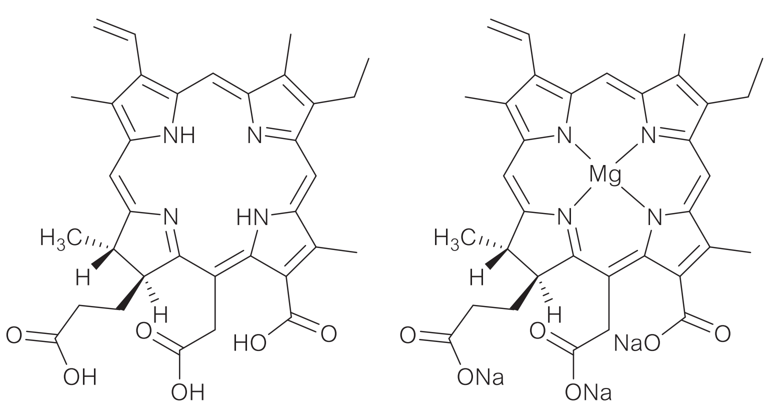


Fig. 3 Chlorin e6 (left) and sodium magnesium chlorophyllin (right).[56]

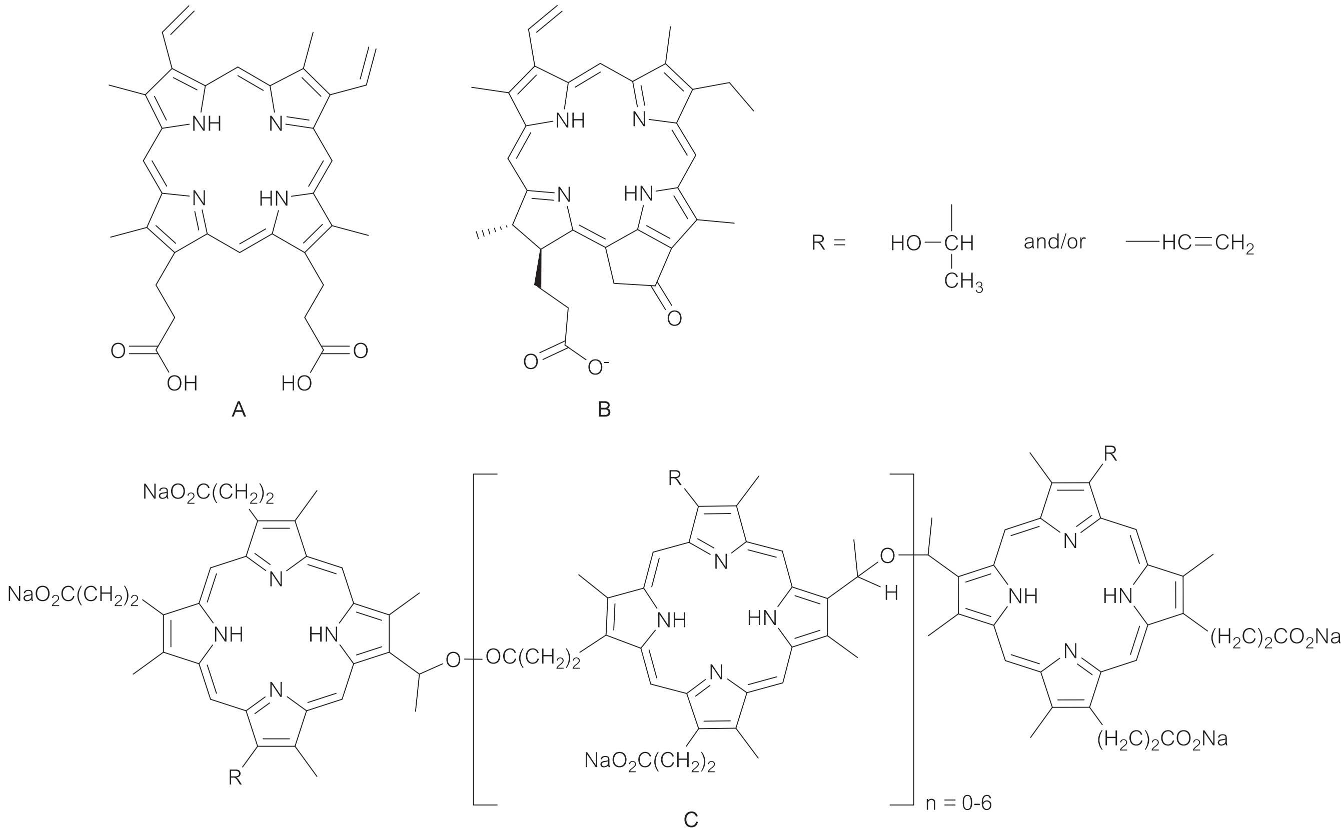


Fig. 4 Protoporphyrin IX (A), pyropheophorbide-a (B), and photofrin (C).[58]

Organoboron compounds (and in particular, boronate esters) are applied in a wide range of transformations, so the development of smooth protocols for the preparation of such compounds is of great interest in synthesis. An excursus on photoinduced borylations, ranging from early stoichiometric photochemical coupling reactions of R(Ar)–H derivatives with metal–boryl complexes to photoredox catalyzed borylations have been showed by Tian *et al.*[59] Although transition metal-catalysed asymmetric coupling is considered a well-established protocol for the regio- and enantioselective formation of stereogenic centres, the adoption of approaches merging cross coupling and photochemical/photocatalyzed processes has received attention only recently. The advancement in this field has been summarized in term of substrate scope by a tutorial review of Xiao and co-workers (an example in Fig. 5).[60] Several examples of visible light mediated radical coupling reactions catalysed by ruthenium, and iridium complexes as well as by organic photoredox agents in the 2011–2021 period have been presented by Bell and Murphy.[61]

It is well known that the choice of solvent has a key influence on the outcomes of a photochemical process, since solute – solvent interactions modify the energies of and crossings between electronic states of the chromophores. Venkatraman and Orr-Ewing resumed their recent efforts in investigating the effects that different solvents has on the dynamic of photoexcited compounds by means of ultrafast laser spectroscopy methods.[62]

The use of a single semiconductor photocatalyst to cause simultaneously hydrogen evolution and the oxidative formation of a C–X bond

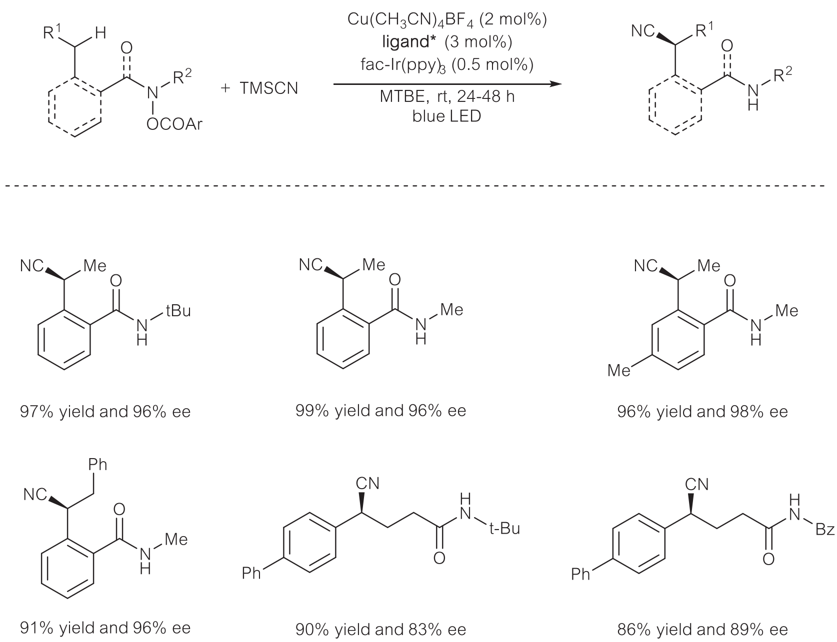


Fig. 5 Copper-mediated photoredox benzylic cyanation.[60]

under visible light irradiation is considered as a challenge in modern sustainable organic synthesis. Such cooperative photoredox coupling of H_2 production and selective organic conversions (including the dehydrogenation of cyclohexane to benzene mediated by Pt@TiO_2 nanoparticles) have been exhaustively reviewed by Qi *et al.*[63] Published as a part of the virtual issue CO_2 *Reductions via Photo and Electrochemical Processes,* (expected publication: 2022) the review of Son *et al.* summarized the most innovative multicomponent inorganometallic (MIOM) hybrid systems employed in CO_2 reduction.[64] R. Godin and G. R. Durrant focused on key role of the charge carrier dynamics in the functioning of photovoltaic devices and photosynthetic systems, pointing out that, since the charge extraction processes in photovoltaic devices is faster the multi-electron/proton redox reactions exploited in photosynthetic devices to produce chemical fuels, the former exhibit a small energy losses.[65] Metal halide inorganic perovskites are currently considered as the elective materials for photovoltaics applications, that allows for the achievement of a power conversion efficiency (PCE) over the 20%. The main loss lies in the photovoltage, which is limited by interfaces in terms of nonradiative recombination. This review of Tress and co-workers summarizes the recent efforts in investigation how the interfaces and interfacial layers affect the performance of solar cells based on inorganic perovskite absorbers, proposing some paradigms for the development of both efficient and stable inorganic PSCs.[66] Finally, a review on the application of supramolecular strategies to the development of artificial photosynthesis systems has been written by Keijer *et al.*[67]

The noninvasive approach of PDT makes it as the elective alternative to conventional cancer treatment and has been the subject of a myriad of research papers but the rational designs of organic photosensitizers (OPSs) to be used in PDT is an aspect that has been poorly investigated in reviews. The report of J. Yoon and coworkers is a detailed collection of examples of the recently developed OPSs, that have been classified in view of their mode of action (see Fig. 6).[68]

The adoption of nanomaterials, either as carriers or photosensitizers, may overcome the limitations to the clinical application of PDT including poor tissue penetrability of light, tumor resistance to the treatment and thus non satisfactory induction of tumor cell death. Nanocomposites that absorb in a wide range of the light spectrum have been recently proposed in the PDT of deep-seated tumors. In particular, the modulation of cell death pathways with co-delivered reagents enables both PDT induced tumor cell death and tumor inhibition. An exhaustive summary of the efforts in the preparation of nanocomposite photosensitizers (including, among others, noble metal nanoparticles and metal oxide nanoparticles) their clinical application was presented by Xie *et al.*[69] The different strategies used to improve the efficiency of PDT in the treatment of hypoxic tumors, namely the enhance of oxygen concentration in the tumoral cell, approaches to disregard hypoxia and exploiting hypoxic conditions (*e.g.* by the use of hypoxic cell sensitizers such as 2-nitroimidazole or agents bearing an azobenzene moiety, see figure PD) have been presented by Huang *et al.*[70]

Fig. 6 Photosensitizers developed classified in view of their mode of action. Reproduced from ref. 68 with permission from American Chemical Society, Copyright 2021.

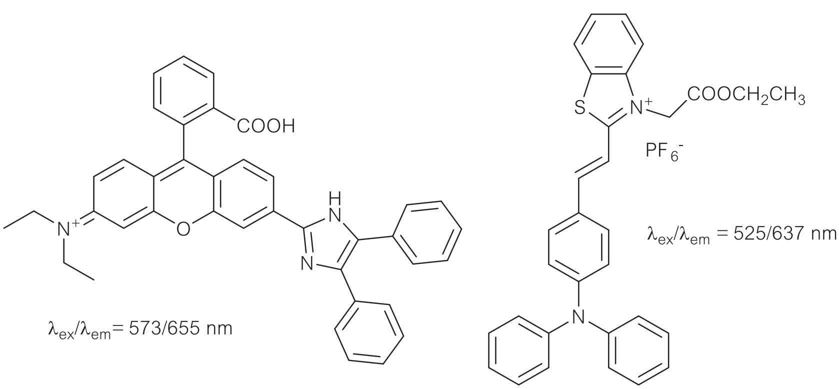


Fig. 7 Examples of fluorescent chemical probes used for intracellular viscosity measure.

A review on fluorescent probes recently developed for the measure of the chemico-physical properties of a cellular microenvironment (*e.g.* temperature, viscosity, temperature, oxygen concentration, and pH, two examples are presented in Fig. 7) has been presented by Yin *et al.*[71] The development of fluorescent sensors based on a twisted intramolecular charge transfer (TICT) mechanism has been reviewed and analyzed by Liu and co-workers.[72]

Acknowledgements

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