



Full paper/Mémoire

Recent advances in artificial photosynthetic systems at Newcastle University

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ABSTRACT

This review covers the most significant research done by the group of scientists working at Newcastle University during the last five years in the area of artificial photosynthesis. In particular, it tackles the aspects of light harvesting in molecular systems and solid state materials, photocatalysis in homogeneous and heterogeneous phases, and the fabrication of photoelectrochemical devices for the generation of hydrogen.

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1. Introduction

This review article aims to show the research developed by the current group of scientists at the School of Chemistry at Newcastle University in the last five years, focused on artificial photosynthesis and covering various aspects from light harvesting and electron transfer to catalysis and devices. The researchers in Newcastle are currently involved in this field and the work hereafter showcased are of Professors Anthony Harriman and Andrew Benniston, founders of the Molecular Photonics Laboratory in 2001, who recently were joined by Dr Pau Farràs (Materials Science Institute of Barcelona and Institute of Chemical Research of Catalonia), Dr Fabio Cucinotta (University of Muenster and University of Eastern Piedmont), Dr Anna Reynal (Institute of Chemical Research of Catalonia and Imperial College London) and Dr Elizabeth Gibson (University of

Nottingham). Although, some of the studies showcased below from the latter four researchers have not been carried out in Newcastle, the authors would like to stress that the experience and knowledge in the field is well represented by the current researchers in Newcastle and will be demonstrated in the following sections.

Artificial photosynthesis (AP) is an old term to describe the area of science that studies the natural processes of photosynthesis and tries to mimic them in artificial systems. Early texts on the topic can be found in Japan and the USSR but were mainly related to plant biochemistry [1]. In the 70s, the oil crisis prompted a huge demand for new and alternative energy sources, and AP was already thought to be a viable and promising solution to help solve the energy issue in a sustainable manner. Most of the work done was summarized in the report of Archer *et al.* [2]. AP became the hot topic from the 70s on and the early studies of Fujishima and co-workers on molecular water splitting using TiO₂ [3a] are seen to be seminal contributions to the field. Since then, most notable scientists, including Graetzel, Lehn, Balzani, Wasielewski, Meyer, Porter and Harriman [3b–g], have been working on this topic. Harriman's early report on

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AP was published in 1978 in Nature [3h], and almost 40 years later, there has not yet been a practical demonstration of economically viable technology.

In Fig. 1 the effect of the oil crisis in 1973 on the number of publications related to AP in the following years is shown. After the first series of preliminary results achieved worldwide with a generally steady publication rate, there has been a steep increase of papers from 2009 onwards. The timescale coincides with the publication of the first single-site molecular system for light-driven water oxidation, which is well known to be the bottleneck for light-driven water splitting [4a], and photochemical-assisted water splitting using a photoelectrochemical cell [4b]. At the same time, the high resolution of the crystal structure of the natural Photosystem II (PSII) helped to improve the understanding of the mechanism of water oxidation in the natural photosystems [5]. Overall, along with the observed increase in the number of scientific publications, the amount of investments worldwide from both the public and private sectors has ramped up. An example of this was the establishment of the UK Solar Fuels Network in which Prof. Harriman, as the Royal Society of Chemistry's Solar Champion, is still actively involved.

In the following sections, the most remarkable work involving current research studies at Newcastle University in the topics of light harvesting, photocatalysis and devices will be summarised, in order to give an overview of the expertise acquired in recent years both in molecular systems and solid state materials.

2. Light harvesting systems

2.1. Light harvesting studies using molecular systems

The study of light harvesting and electron and energy transfer has been the main focus of research within the

photochemistry community in Newcastle. The supramolecular assembly found in natural photosynthesis is a very complex machinery that absorbs light and transfers it to a reaction centre located in the membrane of the plant chloroplasts, where the catalysis takes place. At the same time, the light-driven process is able to redirect some of the energy absorbed to other sections of the membrane in order to protect it from overexposure to light.

Synthetically, one of the main interests has been the preparation of highly fluorescent dyes with the aim of minimizing non-radiative energy losses while maximizing the efficiency of light absorption over the visible spectrum by the dyes. Simple molecules have been studied using standard photophysical techniques including 1*H*,3*H*-isochromeno[6,5,4-*mna*]xanthene-1,3-dione (CXD) [6].

In the natural system, multiple donor-acceptor units are present with uniquely high spatial organization and, taking inspiration from the design of these photosystems, a vast range of multi-chromophoric complexes have been prepared with the aim of studying and manipulating the charge-transfer processes occurring at both intramolecular and intermolecular levels. Boron dipyrromethene (bodipy) dyes have been extensively studied in Newcastle by Harriman and Benniston and are described in the following sections [7].

2.1.1. Small dyad systems

The understanding of charge transfer processes in small systems has been the fundamental for the preparation of more elaborated complexes. Electronic energy transfer strongly depends on the distance between the donor and acceptor and also on their mutual spatial organization. To maximize the transfer efficiencies, cofacial bodipy dyads were prepared with varying lengths of the bridging unit and their transfer rates were measured to give a clear correlation between distance and rates [8]. Nevertheless,

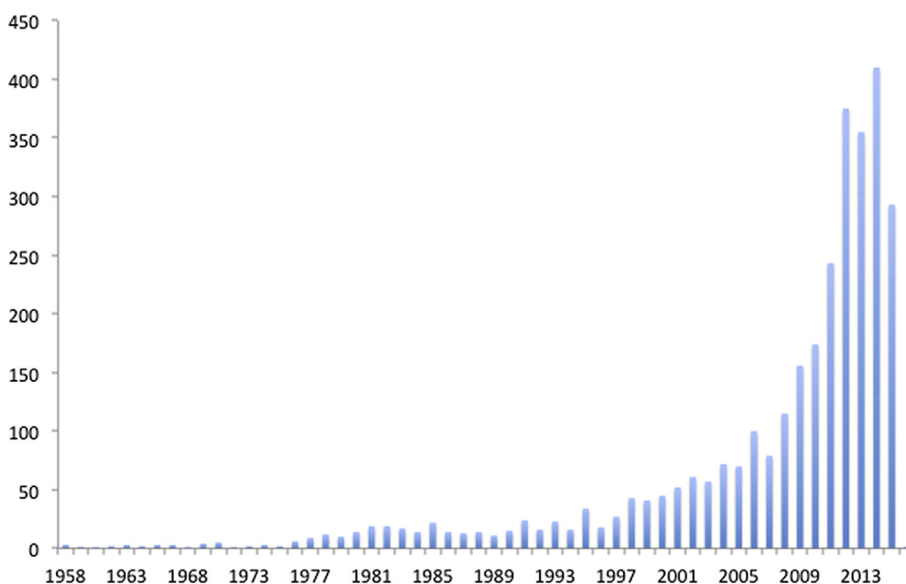


Fig. 1. Evolution of publications related to Artificial Photosynthesis in the last 50 years. Source: Scifinder 2015 (accessed September 9, 2015).

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