Expert View from Science

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Artificial Photosynthesis for Solar Fuels – an Evolving Research Field within AMPEA, a Joint Programme of the European Energy Research Alliance

Abstract: On the path to an energy transition away from fossil fuels to sustainable sources, the European Union is for the moment keeping pace with the objectives of the Strategic Energy Technology-Plan. For this trend to continue after 2020, scientific breakthroughs must be achieved. One main objective is to produce solar fuels from solar energy and water in direct processes to accomplish the efficient storage of solar energy in a chemical form. This is a grand scientific challenge. One important approach to achieve this goal is Artificial Photosynthesis. The European Energy Research Alliance has launched the Joint Programme "Advanced Materials & Processes for Energy Applications" (AMPEA) to foster the role of basic science in Future Emerging Technologies. European researchers in artificial photosynthesis recently met at an AMPEA organized workshop to define common research strategies and milestones for the future. Through this work artificial photosynthesis became the first energy research sub-field to be organised into what is designated "an Application" within AMPEA. The ambition is to drive and accelerate solar fuels research into a powerful European field – in a shorter time and with a broader scope than possible for individual or national initiatives. Within AMPEA the Application Artificial Photosynthesis is inclusive and intended to bring together all European scientists in relevant fields. The goal is to set up a thorough and systematic programme of directed research, which by 2020 will have advanced to a point where commercially viable artificial photosynthetic devices will be under development in partnership with industry.

Keywords: solar fuels, European Energy Roadmap, Joint Programming

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1 Background

1.1 Addressing social goals in energy research

Europe and most of its individual countries depend heavily on outside fossil energy resources. These are dwindling and their supply can easily be jeopardized. In addition, the use of the presently dominating energy carriers based on fossil fuels is connected to environmental and social factors that clearly affect the quality of life. In particular this concerns coal, oil and gas with severe availability constraints, pollution problems and alarming CO_2-emissions. Catastrophes, in particular the damage done by the tsunami in 2011 in Japan, have increased the public worries about large future nuclear power programmes, and in addition significantly increased the projected costs of new nuclear power plants.

1.2 Organisation of European energy research

These factors have prompted the European Union to accept that an energy transition will occur and enter into a planning phase the aim of which is the progressive replacement of fossil fuels by clean energy, mainly from renewable resources. This long term vision has two timescales. In the near and mid-term future, the 27 member states have already set targets for the share of renewable energy in the overall consumption in 2020, with an average of 20% for the entire Union. Enacted as an EU directive [1], this goal is part of the Strategic Energy Technology Plan [SET-Plan]. For the longer term, an energy roadmap has been drawn up all the way to the year 2050 [2]. It calls for a reduction of more than 80% in greenhouse gas emissions, for a strong commitment to enhance energy efficiency and for efforts to increase the penetration of renewable energy to reach all sectors of society. The use of renewable energy is intended to reach levels higher than 50% of the total European energy consumption by 2050.

Today the 20% renewable energy target for 2020 appears reachable, provided that the installation of facilities producing renewable energy continues to increase at the current rate. From 2004 to 2010, the share of renewable energy in Europe experienced an annual growth of about 8% (Figure 1). If this growth continues at the same rate, it has the potential to bring Europe above the target set for 2020 (although this certainly requires substantial

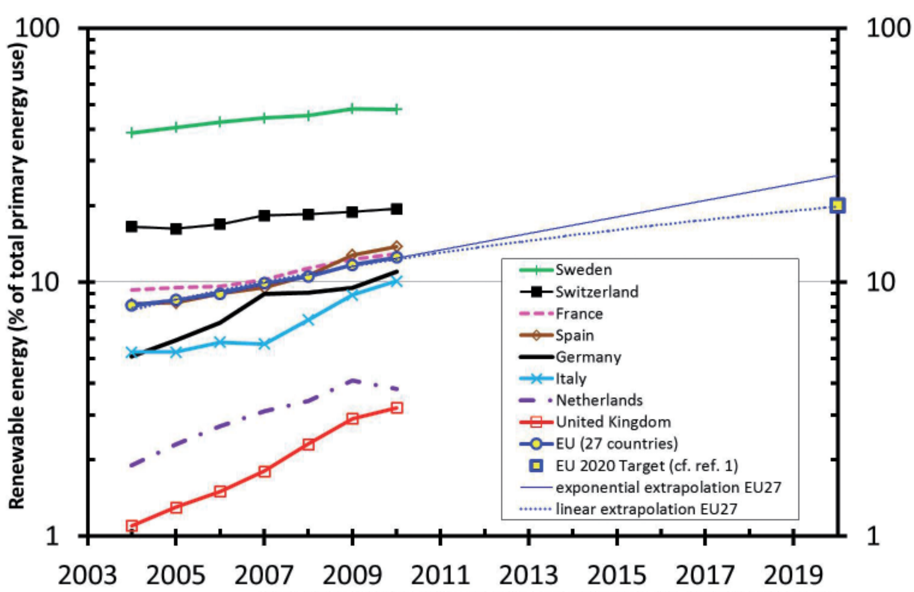

Fig. 1: Share of renewables in primary energy consumption in the European Union (in %) plotted on a log scale – data from Eurostat and from the Swiss federal office of energy for the period 2004–2010. In most European countries (we show here only the statistics for the home countries of the authors of this contribution), renewable energy was promoted from the beginning of the century leading to a steady development. The present pace permits some of the targets defined for 2020 to be reached [1]. After 2020, scientific and technological breakthroughs are needed, especially if renewable energy use is to continue increasing to levels higher than 50% for the whole EU in 2050. For this, the production of solar fuels by Artificial Photosynthesis would represent a key breakthrough. It is time now to invest in an Artificial Photosynthesis roadmap, if we want to maintain or even accelerate the present trend in the energy transition.

financial investments). However, in order to maintain the pace of the energy transition until 2020 and beyond, there is a real need for a major research effort in many energy sectors. These have to address basic and applied topics concerning both the organisation and the technology of the entire energy sector.

Together with the launch of the SET-Plan in 2008, European public research and technology organisations were encouraged to unite their scientific competences: ten founding partners constituted the European Energy Research Alliance (EERA) [3] and started to work at defining and implementing Joint Programmes designed to accelerate research towards low carbon technologies and to rationalise investments. The alliance now includes more than 150 participating institutions throughout Europe. Its raison d'être is threefold: a) to minimize redundancy and duplication in research; b) to concentrate funding on the best performing teams; c) to build trans-national teams of a critical size.

To date, EERA has launched 13 Joint Programmes. Most of them are focussed on wind, solar (thermal and photovoltaic), biomass, geothermal, and ocean energy and the technologies crucial for their implementation (for example smart grids). In particular the Joint Programme "Energy Storage" aims at improving and tuning efficient large-scale storage capacities for grid-connected wind or solar facilities. Chemical storage, for example the conversion and storage of solar energy in the chemical bonds of hydrogen (H_2) made from water, is being increasingly implemented in the so-called "Power-to-Gas" scheme. Considering the 2050 road-map [2], it will with time become necessary to convert an increasingly larger share of the renewable energy into gaseous or liquid fuels. One important reason is that most of the energy consumption in Europe (and globally) is not in the form of electricity, which is the product of wind-power, solar cells, concentrated solar heat and several other renewable technologies. Instead, more than 80% of the primary energy is used mainly as fuels to drive a vast variety of processes in our societies (see e.g. Eurostat) [4]. A second reason is limitations of the electricity grid, which prevents very large penetration of renewable electricity coming from intermittent energy sources often located far away from the final users. One promising solution to both these problems is Artificial Photosynthesis which provides all of the energy supply, the necessary energy storage and the facilitated transportation. In addition, Artificial Photosynthesis allows utilisation of renewable energy at both the local and the continental level producing fuels made from solar energy and water.

1.3 Solar fuels and artificial photosynthesis

The introduction of CO_2-lean, transportable energy carriers made from renewable resources would directly minimize the use of fossil fuels. Unfortunately, biomass-based fuels (although heavily developed and dominating today) can only be important on a restricted scale. Globally there will not be enough biomass to allow for biofuel production to exchange for the fossil fuels. This holds today and even more so in the future when the global energy demand has increased further. Solar energy represents by far the largest of the presently useful renewable energy flows. Therefore, the vision of *solar fuels as important future energy carriers* makes this a central research theme that is quickly moving up the global agenda. The terms "solar fuel", "artificial photosynthesis," and "artificial leaf" have become established in recent years. All of them reflect the same goal, to make a fuel from solar energy and sustainable raw materials, in particular using water as the electron source (Figure 2).

In the long run, new scientific breakthroughs will be needed for solar fuels or other future emerging renewable energy technologies to become used on a scale that is necessary to replace fossil fuels. Basic, multidisciplinary science is required in order to push forward the energy transition by the second half of the century. In 2008, the European Science Foundation published a science policy briefing calling for an action plan towards the development of solar fuels [5]. The ESF later sponsored a EUROCORES programme for solar fuels, which is currently funding two solar fuel networks dealing with some aspects of the scientific road-map to solar fuels [6]. To increase the impact of innovation coming from academic research, EERA recently started a basic science initiative: the Joint

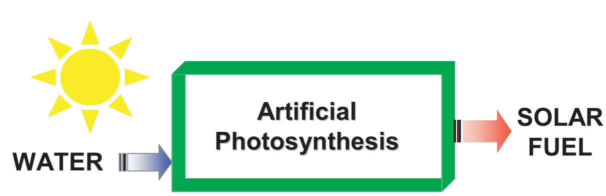

Fig. 2: Overview of artificial photosynthesis. The vision is to produce a solar fuel from solar energy and water. There are different solar fuels but they are always made with solar energy as the only energy source and water as the electron source. The science in AMPEA Artificial Photosynthesis is intended to develop direct energy conversion processes where water is the raw material. The box represents the many challenging research subfields involved. From an applied perspective the box represents the future systems or devices, which will be producing the solar fuel from solar energy and water.

Programme AMPEA, standing for Advanced Materials & Processes for Energy Applications. AMPEA was launched at the end of 2011 and to date more than 40 European scientific institutions participate in the programme. In Parts 2 and 3 of this paper, we will describe how the application of the sub-field "Artificial Photosynthesis" will be developed within AMPEA.

In recent years there have been calls for investments in science in order to obtain breakthroughs on energy technologies. Steven Chu, Nobel Prize laureate in Physics and present Secretary of Energy of the United States, said that on the subject of energy that he was "optimistic that science can offer better solutions than we can imagine today . . . if we harness the creativity and . . . intellectual horsepower of our best scientists in the right way" [7]. The Department of Energy (DOE) organised a consultation of national and international experts which produced orientation documents and eventually gave birth to new funding schemes for energy science. DOE funded a series of Energy Frontier Research Centers of which several have large solar fuels activities. DOE also created the so-called Energy Innovation Hubs in 2010, and among them, a Joint Centre for Artificial Photosynthesis (JCAP) [8] was established between the California Institute of Technology and the Lawrence Berkeley National Laboratory in California. Currently this is probably the world's largest operation (two dedicated laboratory buildings, one at each campus) on artificial photosynthesis purely aiming at the development of solar fuels. In Europe, the Swedish Consortium for Artificial Photosynthesis (CAP) was the first large collaboration of this kind [4]. The Swedish CAP started much earlier, in 1994, and is since 2005 concentrated at Uppsala University which hosts the largest laboratory entirely devoted to solar fuels research in Europe. In Europe several new efforts of this dedicated type are being launched in for example Italy [9] and The Netherlands with the BioSolar Cells consortium [10]. Also elsewhere in the world, especially in Japan, Korea, China and Australia, ambitious programmes have been initiated. This burgeoning activity reflects that solar fuels is one of very few options that mankind can develop to achieve and maintain a sustainable future for the 9–10 billion people anticipated to be living on the planet by the year 2050 [5].

1.4 AMPEA and artificial photosynthesis

AMPEA has a matrix structure which is shown in Figure 3. The science, intended to drive the European energy research in areas where the basic science content is high, will be carried out in energy applications. The applications are crossing three technical sub-programmes (SP) that involve basic scientific tools. For SP1 these are the design and synthesis of new materials; for SP2 the physical modelling of materials, devices and processes; and for SP3 the characterization of materials, devices and processes. The technical sub-programmes constitute a core base and are intended to deepen the research carried out in the scientific applications. Artificial Photosynthesis is the first of these applications. Its creation (Figure 3) within the AMPEA Joint Programme is intended to boost research in this area on a pan-European basis. Its immediate inclusion in the first phase of the AMPEA Joint Programme reflects its urgent character. Artificial Photosynthesis in AMPEA should be inclusive and gather all scientists active in related fields in Europe. This research community is diverse and already quite large. It is rapidly growing by attracting young researchers and the field in Europe is estimated to already involve as many as 100 internationally recognized PI's in more than 15 countries.

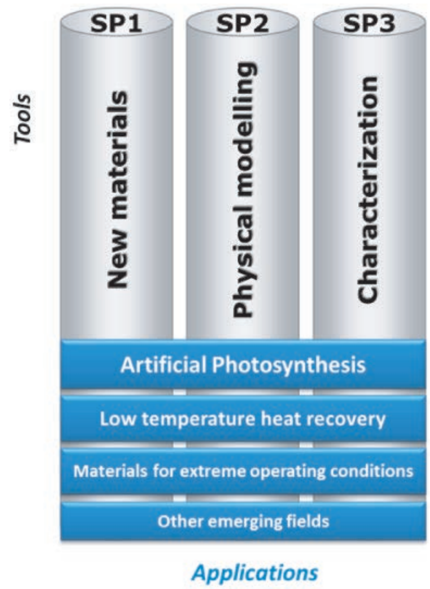

Fig. 3: The structure of AMPEA is a matrix of tools and applications aimed at fostering the development of future emerging energy technologies, while favouring the use of scientific tools provided by researchers of the Applications domains. AMPEA Artificial Photosynthesis is described in the present text. Other emerging applications are currently being defined.

A solar fuel is always made using solar energy as the only energy source. Huge amounts of solar energy are available everywhere and are often abundant, even in densely populated areas. In contrast to other renewable energy options there is more than enough solar energy available to allow the shift from fossil fuels, even at fairly modest conversions efficiencies of the solar energy to the desired product (a solar fuel or solar electricity). Even in

Northern Europe there is enough solar energy to contribute substantially to the energy system in the summer with the long days. However, the solar energy is lacking in winter when energy demand is high. Therefore, solar energy can become a truly important energy source also in these very northern countries only through its conversion into a storable energy carrier, the solar fuel.

For the widespread and cost effective development of solar energy in Europe, it is critical that very efficient processes are developed. In particular, it is urgent to develop high efficiency artificial photosynthesis routes for the *direct conversion* of solar energy into fuel [4, 9]. Such routes are not yet available and the AMPEA Artificial Photosynthesis Application is created specifically to address this point. Indirect processes for fuel production, using for example electricity or biomass as intermediary energy carriers, appear – even though they work – less optimal because of inherent energy losses. They are not part of AMPEA but are studied partially by other Joint Programmes of EERA. Synergies shall be profitably developed between these parallel paths towards solar fuels, on catalysts, electrolytes, materials synthesis, design of devices etc.

The raw material for the fuel is the second, equally important, key aspect of this process. Artificial photosynthesis targets *water as the raw material* [4]. Thus, AMPEA Artificial Photosynthesis gathers basic and applied science with the aim of providing a CO_2-lean fuel based on solar energy and water (Figure 2). We envision extensive development of artificial photosynthesis for solar fuels as a replacement for fossil fuels thereby avoiding the effects on the climate caused by the current extensive use of fossil fuels. The road to success will be long and winding, the stakes are high but the potential benefits enormous. Scientific breakthroughs in this area could quickly be transformed into technology, lead to new industrial sectors, and drive positive societal changes. At this moment the field of solar fuels in Europe is scattered. AMPEA Artificial Photosynthesis is designed to change this.

The intended solar fuel should address the market demand that could vary in different countries or regions. Presently there are only a few ideas for solar fuels that are being pursued internationally. AMPEA Artificial Photosynthesis intends to gather the available science in Europe to design several promising routes for fuel production from solar energy and water [4]. Many scientists target hydrogen as the solar fuel. This is driven by intentions in for example the European road map towards the hydrogen society (in which hydrogen should be non-carbon based by 2050) and is a natural choice when water is the raw material. Proof of concept principles already exists and it is possible to produce hydrogen, even at high pressure, with artificial photosynthesis. However, other solar fuels, such as carbon containing solar fuels like alcohols produced from concentrated CO_2 sources, are also potential targets [9, 11].

A key element in the research is *solar energy conversion*. AMPEA Artificial Photosynthesis aims to use light energy to drive all reactions leading to fuel formation and to develop artificial photosynthesis in supra-molecular systems and in photo-electrochemically active nanostructured materials. Molecular sensitizers or semiconductor materials will be employed to harvest the light and to convert the energy into an electrochemical potential able to drive electrons between catalysts. All components have to be made from cheap and abundant elements and the system must drive both the efficient oxidation of water and reductive processes (hydrogen evolution and/or reduction of carbon dioxide) to generate the fuel. A core activity to achieve this is the design and synthesis of stable, cheap and efficient catalysts for water oxidation and fuel production. A central feature of such catalysts is their ability to perform advanced electron and proton management. Advances in this field will benefit from suppress by detailed biochemical, structural and biophysical knowledge of enzymes that carry out these processes in nature (hydrogenases, water oxidizing enzyme and photosynthetic reaction centres) and biology will also serve as inspiration in this field. The integration of the knowledge from functional natural systems into man-made catalysts is a key aspect of this programme.

Energy relevance – strategic objective. The strategic objective of AMPEA Artificial Photosynthesis is to meet demands for an exchange for fossil fuels by the production of solar fuels from solar energy using water as the electron source. When this is accomplished, solar energy will be *converted* to chemical energy in a fuel. The fuel can be *stored*, from summer when solar energy is abundant to winter when demand is high, from daytime with strong sunshine to evenings and nights etc. It can be *transported* to where it is needed. It has the potential to meet the present need of ca. 80% fuel in the energy consumption. This research is complex, challenging and without shortcuts. It is consequently long-term and reaches into a time-domain beyond 2020. It demands work in cross-disciplinary constellations of researchers where expertise from many different fields join forces. AMPEA Artificial Photosynthesis addresses an area where Europe has an edge with respect to other international research efforts. The programme will create new interfaces where strong synergies can be anticipated. Breakthrough results and unexpected quantum leaps in the science are expected to be promoted at these interfaces allowing development towards large-scale application through the new collaborations explored in AMPEA. The programme integrates a broad arsenal of scientific techniques and thinking/knowledge from scientists with many competences. The planned research is fundamental but with a strong emphasis on addressing the total conversion chain, by integration of parts and modules to establish functional devices, first on the laboratory scale and later on the industrial scale.

AMPEA Materials sub-programme SP1 aims at rationalising routes of materials synthesis, and materials assembly in the framework of nanotechnologies: for Artificial Photosynthesis, benefits will concern the concept of electrodes and membranes, the integration of catalysts properties and the optimisation of interfaces that are main sources of energy loss. An important feature in AMPEA is the merging of materials science with the study of physical and chemical processes within the modelling and characterisation sub-programmes SP2 and SP3. The complex multidimensional problems of energy solar fuel producing devices will be investigated in specific platforms, by developing specific modelling tools and by using dedicated large-scale instruments. In situ operando characterisation will give insight into the materials and processes at the heart of the photo-catalytic devices designed for artificial photosynthesis.

2 Objectives

2.1 Long-term impact: a conceptual change of today's fossil based energy system on the terawatt scale

2.1.1 Energy source and raw material

Solar fuels are made using *solar energy as the sole energy source and water as the raw material*. They will consequently be able to provide CO_2-lean, renewable and storable energy carriers in almost unlimited quantities. Today, functional and cost-efficient direct technologies for solar fuels production are not available and the necessary science to change this is the focus of AMPEA Artificial Photosynthesis. The long-term vision is that the development and use of solar fuels shall become a major factor to alleviate the effects of the extensive use of fossil fuels, thereby providing a paradigm shift away from our present fossil-based energy system.

For 1^{st} or 2^{nd} generation biofuels (the presently dominating renewable fuels), biomass or waste is the raw material. While these biofuels might be very useful in certain regions or countries, they are always limited by the availability of the corresponding raw material. This limits their application on a global scale and such methods are *not covered* in this programme. Instead, the intended raw material must be essentially inexhaustible, cheap and widely available [4]. There are few options and *the target in this proposal is to develop processes where water is the raw material*. The question of the availability and cost of clean water can be raised. However the water usage in processes for solar fuels production using artificial photosynthesis is orders of magnitude smaller than for agricultural processes [12]. The amount of water used in the process is therefore of little concern from an environmental perspective. Processes where water is split into its constituents by solar energy can become large contributors to shift away from fossil fuels on a global scale. The development of technologies for direct solar to fuel conversion requires revolutionary breakthroughs in several areas of fundamental science. Equally important is the integration of the different parts and modules in smart (responsive) matrices at an early stage and to produce prototypes and functional devices. An additional mind-provoking possibility is to use semi-artificial components from natural species that are engineered for superior performance beyond what nature has to offer.

2.2 Scientific objectives and linked research areas

AMPEA Artificial Photosynthesis will focus on direct processes for solar fuel production involving several research subfields. It will provide great possibilities for breakthrough research and unexpected, rapid progress. Novel synergies will emerge between the research in the scientific applications and scientists in the technical subprogrammes of the AMPEA Joint Programme.

Figure 4 shows an overview of the science and subfields in AMPEA Artificial Photosynthesis and Figure 5 illustrates a few representative scientific details from the respective subfields. For a long time, the most critical hurdle to be overcome has been the design of a stable, light driven catalyst that can oxidise water at sufficient rates. The catalysts for water-oxidation and fuel production can be *molecular and/or solid state or nanomaterials*. They share similar physical principles and similar, major scientific challenges are encountered in multi-electron catalysis and proton-coupled electron transfer. For both molecular and nanoscaled catalysts, water-oxidation is a major challenge. To make the huge quantities of solar

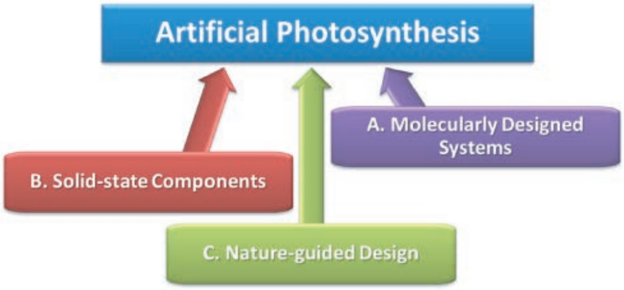

Fig. 4: Overview of the broad scope of the three subfields in AMPEA Artificial Photosynthesis. Numerous breakthroughs probably lie in the collaboration between the three subfields, in the design of hybrid devices as well as in the understanding of basic principles of water oxidation, hydrogen production, light absorption etc. Knowledge transfer from biological systems that function extremely efficiently is a key element in AMPEA Artificial Photosynthesis. The development of the necessary breakthrough concepts will be facilitated by the connection of Artificial Photosynthesis to the technical sub-programmes that bring a wide range of scientific methodologies; abilities to optimize design and synthesis etc. (see e.g. Figure 3).

fuels needed for a transition from fossil fuels it is necessary that the catalysts are made from earth abundant elements like manganese (in natural photosynthesis) [13], iron [14] or cobalt [15] while the use of scarce and expensive elements such as ruthenium, platinum or iridium otherwise might become a prohibitive bottleneck [16]. There are few efficient molecular catalysts based on these abundant elements and development of such catalysts is therefore in the early focus for many research efforts of the AMPEA participants. However, there exist heterogeneous catalysts based on cobalt [15, 17, 18] manganese [18, 19] or iron that are ready to try in first-generation device-oriented research. Such attempts will allow fundamental, in-depth understanding of physical and chemical processes in working artificial photosynthesis devices. The same arguments also hold for available efficient molecular catalysts based on ruthenium which are useful model systems both for basic and applied research [20–22]. To consolidate our basic knowledge and to accelerate the research, we will not wait until the last generation of catalysts are developed. In all concepts, the fuel will be produced utilizing the reducing equivalents made available from the light-driven oxidation of water. The fuel can be hydrogen obtained by the reduction of protons, but also the reduction of carbon dioxide (CO_2) can yield fuels such as alcohols. The latter might be easier to use with current technology but is probably more difficult to make in a direct process (although for instance methanol can be efficiently made from hydrogen in industrial processes).

New frontier science is created in AMPEA Artificial Photosynthesis by linking the *three subfields* overviewed in Figures 4 and 5:

- *A. molecularly designed systems*
- *B. solid-state components*
- *C. nature-guided design*

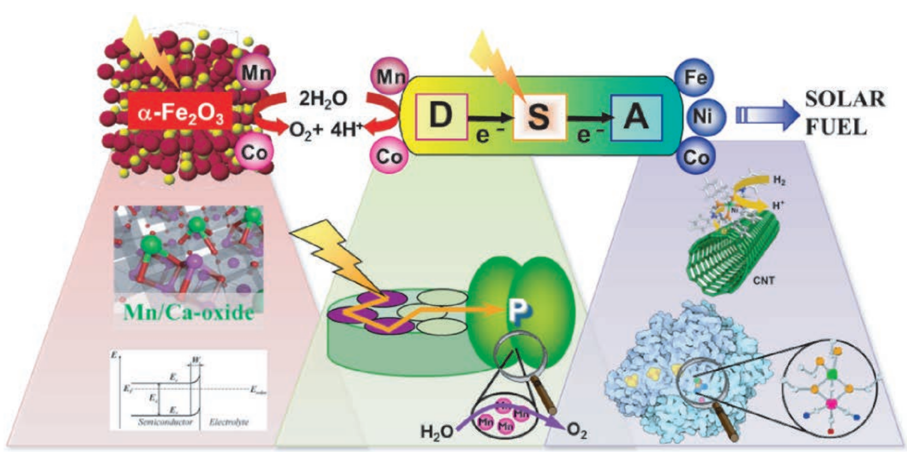

Fig. 5: Overview of AMPEA Artificial Photosynthesis high-lighting the cross-disciplinary nature of the research. Upper part: the solar fuel forming subfields. Right: artificial photosynthesis in devices that make a fuel from solar energy and water using light-absorbers (S) linked to light-driven molecular catalysts made from Mn, Co, Ni or Fe. Left: man-made nanostructured photo-electrochemical systems, here symbolized by hematite (iron-oxide) and a Mn/Ca layer structure used in promising modular assemblies. Lower half: The science stands on solid ground and is built on studies of photosynthetic reaction centres and light harvesting complexes (middle – Photosystem II with antenna and water oxidizing complex), hydrogen metabolizing enzymes (right Ni-Fe hydrogenase). Strong (bio)physical knowledge of solar energy conversion is a key element to the programme. A catalytic system for proton reduction based on carbon nanostructures with a linked catalyst is also shown. Shaded triangles symbolize overlapping areas created in AMPEA Artificial Photosynthesis by synergies with the technical sub-programmes. Some of these will be subject for projects in future calls.

All have strong synergies with the technical sub-programmes (SP 1–3, Figure 3) thereby making AMPEA Artificial Photosynthesis an ideal starting point for immediate activities in application-oriented basic research on renewable energy. In particular, the joint efforts of the field will strive to demonstrate the first generation of viable devices for technical application within the time-frame of the Horizon 2020 framework programme [23].

2.2.1 Sub-field A

The first subfield concerns the use of *molecularly designed systems* (Figures 4 and 5, purple, right). The catalysts are often developed following bio-inspired approaches, but might also contain abundant elements not used by biological systems [24]. The four-electron oxidation of water is clearly one of the main research challenges and a bottleneck for the successful development of artificial photosynthesis. For water oxidation, manganese containing catalysts [25–27] mimicking key aspects of the water oxidizing chemistry in the photosynthetic enzyme Photosystem II [13] and recently introduced cobalt catalysts [15, 17, 28, 29] are promising tracks to follow. There exist also efficient ruthenium catalysts for water oxidation. Although based on a rare element, these are useful to study to increase our knowledge about possible water oxidizing mechanisms and also for early device-oriented work. Two-electron molecular photo-catalysts for hydrogen formation will be based on abundant, cheap transition metals (instead of rare noble metals like platinum) [30]. Biomimetic di-iron and nickel-iron systems mimicking the active site in hydrogenase enzymes [14] are promising but, there are also very useful cobalt-based catalysts [15, 31]. Additionally, the development of stable dye systems for light-harvesting and sensitization are important issues which need attention and evaluation [20, 30]. Within the time-frame for Horizon 2020, the research in this subfield will develop second-generation devices for technological evaluation.

A strong advantage with molecular catalysts is that it is easier to achieve very high degrees of mechanistic understanding than in other conceivable systems. The molecular catalysts can be rationally tailored, tuned and studied for mechanism and performance using a multitude of sophisticated spectroscopic techniques, many available within the technical sub-programmes of AMPEA. This is necessary both for an incorporation of molecular catalysts and dyes into functional devices and also for the design and development of next-generation components.

2.2.2 Sub-field B

For the developments of *solid-state components* (second subfield; Figures 4 and 5, red left) AMPEA Artificial Photosynthesis will investigate solar-driven hydrogen production via photo-electrochemical water splitting [32, 33]. Thin-film photovoltaic materials can be envisioned as short or medium-term solutions if corrosion issues can be solved through the development of protective mechanisms or passivation layers [34]. However, the long-term target should be to construct a cheap nanostructured semiconductor device based on earth-abundant elements [35]. Novel design strategies for the coupling of inorganic components to catalysts need nanotechnology and material science to prepare innovative composite structures in which each component performs its specialized function. Again, one of the key issues in this context is the design of *catalytic nano-particles* or incorporation of *molecular catalysts* that will be interfaced with semiconductors to catalyse water oxidation and hydrogen (fuel) formation. High surface areas to maximise the contact between the catalyst and the substrate and low overvoltages for the desired reactions are required. A critical factor here is the interaction with the technical sub-programmes within AMPEA regarding the scalable production of catalysts and structured components. This also holds for establishment of new methods for elucidating elementary steps of catalysis on solid-state surfaces.

A promising concept is the development of mixed systems, for example by combining solid-state scaffolds with molecular light-driven catalysts. These should preferentially be based on abundant materials like iron, manganese, nickel or cobalt. This development will be initiated and facilitated by the close interaction of the researchers within AMPEA Artificial Photosynthesis.

2.2.3 Sub-field C

The third subfield involves *nature-guided design* (Figure 4 and 5, green middle) where we learn from Nature in our attempts to make entirely man-made chemical systems. Knowledge transfer from efficient photosynthetic systems and enzymes suppress and will strongly fertilize AMPEA Artificial Photosynthesis. This subfield also includes several aspects concerning *semi-synthetic/hybrid systems* involving both biological and man-made components [36, 37]. AMPEA Artificial Photosynthesis invites also this science to be part of the initiative. Importantly, knowledge gained from biological principles and the nature-guided design of devices is envisaged as a driving force and inspirational source also for the research teams with no biological background. The understanding and learning from the chemistry of the natural oxygen evolving complex [28] and hydrogenases [38], for example concerning issues with electron and proton management at the catalytic centers [39–41], is of key importance. These processes are poorly understood so far in the context of materials for artificial photosynthesis. The very high quantum yield observed in natural systems may involve proteins as smart (responsive) matrices which reflect the tight coupling between the chemical reaction and the protein motions [42]. The transfer of principles from biological systems to artificial materials and devices will be an important task of the subfield nature-guided design.

2.3 Devices, demonstrators, prototypes – beyond the state of the art

Many necessary chemical building blocks like catalysts and charge separating ensembles have become available in recent years. Several are made and described by future participants in AMPEA Artificial Photosynthesis. The great challenge in AMPEA Artificial Photosynthesis, and its central research goal, is the assembly and optimization of the different parts to construct half-cells and thereafter functional integrated modular systems to assemble complete photo-catalytic devices that can operate with good yield against wasteful back reactions. Few systems like this have been constructed so far [9, 43] but success here would be ground-breaking for solar fuels production. This core element is virtually lacking from earlier research on artificial photosynthesis.

The idea in AMPEA Artificial Photosynthesis is to bring together many existing pieces from various European groups and laboratories and combine these in different device concepts. The aim is to directly connect photosensitizers to catalysts and surfaces or catalysts with semi-conductor materials and to study the half-cells constructed in this way using theoretical, chemical, physical methods as well as implementing advanced engineering knowledge for the first time in this part of the field.

Since there are many different components to choose from (many catalysts, different surfaces and matrices, many photosensitizers) many concepts will be possible and can be explored in such a programme on a European scale. This is the target of the programme and the intention is to have a significant number of laboratory scale pilot devices in operation just a couple of years from now. At this stage it is not useful to make bold promises or define exact goals for efficiencies, durability, costs etc. since the basic science content in the work is by necessity high. However, all work must be pursued with such targets in mind and a fruitful development in the field would be that such targets could be reliably introduced towards the end of the Horizon 2020 programme period.

2.4 Milestones

The core milestones for the AMPEA Artificial Photosynthesis are: i) To build up a joint continental network in artificial photosynthesis research, involving a wide range of sciences with a common goal. ii) To accelerate and focus the European science in this challenging field with global impact more efficiently than can be achieved by national or individual efforts. iii) To prove the principle in a series of device concepts on a time scale of a few years. At this stage, all sub-fields are equally important and each sub-field (Figures 4 and 5) has internal, traditional scientific milestones.

2.5 Synergy between programme elements

The science in AMPEA Artificial Photosynthesis has a broad scope. All methods depend on advanced chemistry at modest temperatures and mild conditions. Larger physical scales than molecular interactions are also crucial in the control and long term durability of the devices. It is anticipated that much progress will be achieved at the new interfaces created between the scientists in AMPEA Artificial Photosynthesis working together and creating new research interactions with scientists in the technical sub-programmes of AMPEA. Multiscale integration and multifaceted understanding are the main goals of the technical sub-programmes. The central goal for Artificial Photosynthesis is the development of functional half-cells or entire solar-fuel cells where for example molecular catalysts are merged with semiconductor surfaces or photoactive material surfaces. Studies of this kind of devices demand interactions between scientists that seldom have collaborated but have complementary expertise. A required target for the entire scientific field of artificial photosynthesis is to address the challenge to provide renewable energy on the terawatt (TW) scale. This will only become possible to reach if the new synergies created within AMPEA Artificial Photosynthesis can be developed and fully exploited. This involves joint workshops between subfields where synergies are emerging or already strong. A useful development would be the creation of a European conference line. This could be timed to years when the Gordon research conference on solar fuels is not held.

3 Roadmap for an artificial photosynthesis network

Artificial photosynthesis has already been the subject of a few seminal European networks (e.g. Ru-Mn for artificial photosynthesis, SOLAR-H, SOLAR-H_2, Solhydromics, nanoPEC, EuroSolarFuels, ArtipHyction, PERSPECT-H_2O, H_2-NanoSolar etc.) and initiatives in several countries provide a fertile ground for further development. To strengthen this important research field on the European level, knowledge from all European countries was brought together on the occasion of a workshop in Mülheim/Ruhr in Germany (October 2012) to achieve the long-term goal of mastering artificial photosynthesis. Their combined expertise at this workshop covered so diverse fields as biochemistry, molecular and supramolecular chemistry, photo-catalysis, photoelectrocatalysis, electrochemistry, material science, theoretical chemistry, optoelectronics, engineering etc. The AMPEA workshop gathered most of the important players of this rapidly developing scientific field on a European level.

The outcome of the meeting was: 1) the identification of bottlenecks and the formulation of milestones for artificial photosynthesis; 2) the definition of the scientific content and vision of AMPEA Artificial Photosynthesis; 3) the outline of a European road map to overcome the scientific and technological hurdles on the way to efficient solar energy conversion into solar fuels on a very large scale; 4) the definition of the necessity to fund this road map on a continental scale to a level required to be competitive with related initiatives outside Europe; 5) the foundation of a European Solar Fuels Society intended to be an organisational platform for the field in Europe.

The scientists participating in the workshop discussed the science within different time frames with respect to the time defined by the SET-plan and where the field currently exists in Europe. This is an on-going discussion and the points described below only present a starting point for the creation of a more detailed and broader road map for the development of solar fuels. The next AMPEA plenary meeting in this process is scheduled for June 11–12, 2013 in the Netherlands.

3.1 Short timescale (2013/2014)

There are no quick fixes in solar fuels research and most of the research is long term. Despite this, an important issue on the short time frame is to organize the field in Europe in an inclusive and scientifically broad manner. It is imperative that both large scientific organisations and smaller groupings in single universities or from smaller countries are welcome and can be accommodated in the networks and programmes created. The specific target is the scientific integration of the technical sub-programmes and the Artificial Photosynthesis Application, which began at the Mülheim workshop and shall be furthered in a joint AMPEA workshop in spring 2013. An important outcome will be the creation of the first calls for proposals in Artificial Photosynthesis within Horizon 2020, the next framework programme for EU research funding. It is also necessary to establish with relevant EU representatives the importance of Artificial Photosynthesis for the EU initiative leading to the "third industrial revolution" [44].

Early discernible bottlenecks and thereby core agendas in the research programmes include: improving both molecular and solid state catalyst efficiencies and stabilities, mechanistic understanding of critical components, controlling electron and proton management, efficient light absorption and charge-separation. The science involved here is both fundamental and application oriented.

3.2 Medium timescale (2015 to 2020)

During this time-frame a programme, jointly funded by member states and the European Union, will develop a series of Artificial Photosynthesis devices on a laboratory scale based on our various technologies. These first generation devices will help to identify new and still remaining challenges and pin-point factors to focus future efforts on. These devices shall take advantage of the regular improvement of molecular and materials design, shall initiate new multiscale modelling actions and get retrofit from them, and shall be the subject of thorough investigations with dedicated instruments and lines of large scale facilities.

During this period it will also be important to develop a second target involving integration of molecular catalysts and solid state matrices in devices, as well as to implement the use of nature-guided approaches. This science will cover mechanistic studies, nano-structuring, light management, interface engineering, smart (responsive) matrices, grafting and assembling of modules and is well suited for efforts within the AMPEA Joint Programme.

Other valuable areas to be developed have to deal with standardisation and agreement on how to compare device performances as well as taking initiatives to develop a common scientific terminology for the field through summer schools, conferences and the writing of joint texts.

The most important goals and commitments for 2020 shall be the availability at this date of methods and devices that industry can begin to consider for a commercial deployment.

3.3 Long-term (2020 and beyond)

At this time the field will have matured and there will be more scientists and approaches involved. The research under Horizon 2020 will enable a strong interaction between academic science and industry and thus allow the development of technologically more advanced devices, testing and evaluation towards commercialisation and life-cycle analyses. It will be possible to approach the 10%-efficiency goal for the conversion of solar energy to fuel. The real target is the development of efficient, long-lasting and cost effective device concepts for solar fuels such that direct conversion of sunlight into CO_2-lean fuels is an accepted and viable part of a future energy mix in Europe. Parallel to this the basic science is now coupled to strong efforts in application oriented research and development driving the first generation of real solar fuel devices towards up-scaling for commercial competitiveness.

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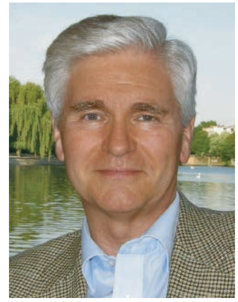

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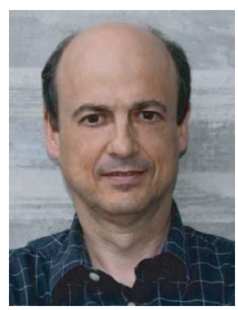

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