



## Review

## Nanostructure in energy conversion

Saskia Buller<sup>a</sup>, Jennifer Strunk<sup>a,b,\*</sup><sup>a</sup>Max-Planck-Institute for Chemical Energy Conversion, Mülheim Ruhr, Germany<sup>b</sup>Center for Nanointegration Duisburg-Essen, University of Duisburg-Essen, Germany

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## ABSTRACT

Nanostructured and nanosized materials are widely applied to tackle the pressing challenges associated with energy conversion. In this conceptual review, rather than highlighting separate examples, we aim to give a general overview about where and how nanostructure design can be beneficial in the three major research fields (photo)thermal chemical energy conversion, electrochemical energy conversion, and solar energy conversion. It will be shown that in many cases the design of catalytically active nanostructures is the main task and that especially for catalysts nanostructure and activity are inseparably linked to each other. Moreover, electrochemical and photochemical processes are complicated by the overlap of multiple processes that all need to be optimized, including in particular light absorption, charge migration, recombination and trapping events and surface processes. It will also be shown how the development of materials for new challenges can often be based on our knowledge on existing materials for related applications.

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## 1. Introduction

It becomes more and more recognized in wide parts of the world that the current system of energy conversion and storage will not be able to last forever. In particular, the main foundation of today's energy infrastructure is based on fossil fuels, namely crude oil, natural gas, and coal. Although the predicted time scales of depletion of the one or other resource might differ as a function of the employed model [1–5], e.g. considering “physical” vs. “economic exhaustion” [1], it is undoubted that alternative solutions will be needed at some point, and potentially sooner rather than later [3–5]. Since uranium is not a renewable resource, either, and since nuclear power has become rather unpopular, in particular due to health and safety issues, nuclear power will likely not be a viable option for the long term [5–9]. Available renewable resources from geothermal energy, hydroelectric or wind power will likely not be sufficient to fulfill the world-wide energy demand; even if their combined full potential is exploited [10]. The only sufficiently available resource we can certainly rely on for the upcoming millions of years is the sun. The energy of the sun reaching the earth's surface is certainly much greater than the recent average annual world-wide consumption, and the factors reported for the excess of the irradiation energy of the sun are ranging between almost six thousand [11] to about four order of

magnitude [12,13]. Or, in other words, if we were able to convert and store all solar energy reaching the surface of the earth in one hour, we would meet the world-wide energy demand of a whole year [4,5]. However, for this purpose we still need to develop efficient tools and processes that allow harvesting and storage of solar energy in a practical form [4,5]. Many approaches are currently in development that are partially linked to each other, but partially go entirely separate ways. Energy conversion could take place, for example, from photons to current (photovoltaics), from photons to chemicals (biotechnological processes or photocatalysis), from photons to heat and/or chemicals (solar thermal approaches), from heat to electricity (thermoelectrics), from electricity to chemicals (electrolyzers) and in certainly many more ways. Energy might then be stored, for example, in forms as different as potential energy in water reservoirs at high elevation, electrical energy in batteries, or chemical energy in chemical bonds of molecules with high energy density [5]. There are scientific approaches in all of those directions, and this is also strongly advised, (a) because we do not know yet which scientific breakthroughs are still achievable in any of those fields and (b) because at present we rely on a mix of energy conversion technologies, too, which demonstrates that different technologies might be needed in different areas of the world and different segments of industry and production.

At the same time, in recent years, there has been much effort to reach further and further into the “nano” domain and to ever smaller sizes, down to the isolated atoms. It is the aim to manipulate materials on their most elementary scale, that is, in their general atomic composition and the arrangement of their smallest

\* Corresponding author at: Max-Planck-Institute for Chemical Energy Conversion, Mülheim/Ruhr, Germany.

E-mail address: [jennifer.strunk@cec.mpg.de](mailto:jennifer.strunk@cec.mpg.de) (J. Strunk).

building blocks [14,15]. Furthermore, sizes and shapes of certain crucial building blocks are made smaller and smaller, because properties might be achieved otherwise not observed in the parent bulk material. Manipulating materials on the nanoscale also requires tools and methods that allow monitoring the achieved manipulation on the nanoscale, if possible under conditions as close as possible to those in the targeted field of application. The high importance of such studies of the working systems, “in situ” and “in operando”, originates from the realization that a structure once established in a particular biological, physical or chemical synthesis not necessarily persists under reaction conditions [16,17]. Often, not just one material is modified and manipulated on the nanoscale. Sometimes, the peculiar properties are a consequence of interfaces and contacts established between different (nano) materials. Those materials, often referred to as “hybrid nanostructures” [18–20], might not only display the properties of their separate components, but also new features not observed in the separate building blocks.

As can be seen from the introduction above, both the terms “nanostructure” and “energy conversion” are much discussed topics nowadays. At first glance, though, they appear entirely unrelated to each other. To the general public, the undoubtedly “macro-sized” solar panels, wind parks, power plants, or catalytic reactors, do not appear to have anything to do with “nano” at all. Yet, it is often the smallest units of a material that make all the difference. Regardless whether the aim is the development of more efficient solar panels, more active catalysts, or longer lasting coatings or electrodes, it has been recognized that modern tools for material modification and manipulation on the nanoscale might help in finding the solution [21]. For example, in the field of heterogeneous catalysis, the possibly most prominent example for the importance of nanosize or nanostructure is catalysis by the noble metal gold: It is well known that bulk gold is chemically absolutely inert; this is a property that has been exploited in the history of humankind to create artifacts that lasted for centuries. On the other hand, reducing the size of gold to that of clusters of a few nanometers and depositing those nanoparticles on oxide supports leads to the formation of a material with high catalytic activity, for example in selective and total oxidation reactions [22–28]. In the field of photocatalysis, the natural oxygen evolution site is the best example for the high importance of nanostructure: Only the one specific  $\text{Mn}_4\text{O}_5\text{Ca}$  cluster with its evolutionary developed protein matrix is active for the oxygen evolution, and so far attempts to replicate such a system (bio)chemically have failed [29,30].

Since the research fields of nanostructured materials and energy conversion are wide enough that books have been written about them (see for example, refs. [31–37]), we have to set certain limits for the scope of this review. With respect to “nanostructure”, we will focus on *inorganic* materials. Undoubtedly, biological and organic materials have an important role in the world-wide energy system, in particular natural photosynthesis. However, in biology and to certain extent in organic chemistry the nanostructure appears to be rather inseparably linked to the chemical composition, i.e. the type of protein or amino acid attached to a certain site or chain. In inorganic materials, on the other hand, shape, size and composition can often be varied in a wide range more or less independent of each other. Furthermore, we will address energy conversion only in the sense of “renewable energy”, thus excluding fossil fuels and nuclear power. Renewable energy *conversion*, however, cannot be addressed without at the same time discussing the issue of energy *storage*, too, the latter often being the more pressing problem [38]. In this review, energy storage will only be addressed with respect to chemical bonds in molecules with high energy density, i.e. by means of the synthesis of so-called “solar fuels”. It appears most reasonable to split the diverse topic of energy conversion into the following sub-topics:

- (Photo)thermal chemical energy conversion
- Electrochemical energy conversion
- (Direct) solar energy conversion

There is certainly significant overlap between all three subtopics. As an example, one might consider a hypothetical photoelectrochemical cell operating at elevated temperatures and under solar light irradiation, in which  $\text{CO}_2$  is converted to methanol. This device would clearly fall into all three categories. Consequently, the following introduction to the three topics will set the boundaries and clarify the definitions that we have adopted in this review.

In the field of (photo)thermal chemical energy conversion, emphasis is clearly put on the production of substances that can be, or at least bear the potential to be, used as fuels on the large scale. Furthermore, in this part of the review, we will only highlight processes that can already be operated on the industrial scale or appear to reach this stage in the near future. Typical processes in this category are the synthesis of methanol or long-chain hydrocarbons from carbon dioxide or the production of liquid fuels from biomass. Consequently, this part of the review will focus for a large part on the properties and the development of (heterogeneous) catalysts, since most of the processes mentioned above require a certain catalyst with well-defined properties. In the field of (photo)thermal chemical energy conversion we will show that the challenges in this regard are often associated with the changing raw material supply, and the drive towards a use of carbon dioxide as feedstock instead of fossil-derived synthesis gas. It will become clear that a thorough understanding of today’s large scale catalytic processes is the best basis to employ nanostructuring technology successfully and fruitfully to develop improved catalyst systems for new challenges.

In the part of electrochemical energy conversion we will highlight the development of suitable, stable and cost-efficient electrode materials and electrocatalysts. Since electrochemical cells combined with photovoltaics currently appear to be a suitable solution for larger scale solar fuel production, i.e. for the splitting of water to generate hydrogen, the development of active and stable catalysts and electrode materials without the use of noble metals is of prime importance. The latter would have little potential for large scale application to satisfy world-wide energy demand. The development of those materials should ideally be based on a thorough understanding of the underlying processes, so the necessary (elementary) reactions associated with the electrocatalytic splitting of water will also be discussed in this section of the review. A combination of manganese oxides as catalysts and carbon materials as electrodes appears to be a promising solution, because those materials are all cheap and abundant. Moreover, a wide variety of nanostructuring approaches to increase the performance are available for these materials, which will be particularly highlighted.

The two most important devices to be considered in the field of solar energy conversion are photovoltaics and photocatalysts. In this field, the main definition is based on the direct use of (sun) light to initiate a reaction, that is, either the generation of a current (photovoltaics) or the synthesis of chemicals (photocatalysis or photoelectrochemistry). In this part of the review, we will highlight the influence of size and structure of a material on properties such as light absorption, charge carrier mobility or the lifetime of photogenerated charge carriers, as those unifying concepts are needed in both the development of materials for photovoltaics and for photocatalysis. In the field of solar energy conversion, it will be shown that many concepts for efficient absorption and conversion of solar energy are already known. While those approaches have often already found their way into application in photovoltaics, the use for photocatalysis is often still being explored. In spite of all the research efforts, the complex interplay between

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