A VISION FOR SUSTAINABLE CHEMISTRY PRODUCTION

- White Paper -
Acknowledgements

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**Introduction**

Modern life is sustained by an unremitting stream of energy that is delivered to final users as fuels, electricity, and heat. Currently, over 80% of the world’s primary energy supply is provided by fossil fuels carbon sources (coal, oil, gas). For the last two centuries fossil fuels, generated from biomass over millions of years, have been extensively used in anthropic activities. When we burn fossil fuels, we liberate the solar energy stored millions of years earlier in chemical bonds, but we are also generating CO\(_2\) as waste. Over the last few decades it has become clear that the CO\(_2\) that is released in this way is affecting the climate stability of the biosphere. Therefore there is a need for an energy transition from fossil fuels to non-fossil-based energies. This transition has already started and must be completed during the present century. The sun gives us an opportunity to complete this energy revolution as it delivers the same energy to the Earth in about one hour as we currently use from fossil fuels, nuclear power and all renewable energy sources combined in a year. Yet, sunlight is a dilute form of energy. It needs to be converted into other forms of energy in order to be used in a profitable way, such as heat, electricity, or fuels.

Nature stores solar energy in carbon-based chemical compounds that mankind has long used as an energy source. In plants, algae and some types of bacteria, solar-driven systems are capable of converting carbon dioxide and water into C\(_2\)H\(_2\)O-containing compounds. Photosynthesis is a way to capture solar energy and store it in the form of chemical bonds. The compounds thus produced can be used as fuels, but also (bio-)materials or chemical feedstock. Developing artificial systems on an industrial scale that generate such compounds from the same sources (i.e. solar radiation, water, CO\(_2\)) is a great challenge. Ideally, such man-made, ecologically friendly energy systems should be more efficient than their natural counterparts, to keep pace with our consumption while making our energy utilization sustainable.

At present, the chemical industry vitally depends on fossil carbon resources, which are both its main energy source and feedstock. Several production processes are very energy-intensive and have low energy efficiency. For instance, nearly half of the fossil oil input in a refinery is used to provide energy. The chemical industry also relies on oil, and to a smaller extent on coal and natural gas, as a feedstock for carbon-based products. Irrespective of the transition to non-fossil sources for our energy systems, humans still require a vast range carbon-based products for diverse use. Preserving fossil carbon resources for non-energy use would therefore be wise. However, finding a way to recycle carbon by converting CO\(_2\) into chemicals, materials or fuels would be even wiser. Although nearly any energy source can be used to drive chemical reactions, sunlight is rarely used for this purpose directly. **The objective of this White Paper is to show that it is possible, and even necessary, to drive chemical reactions by the energy from the sun in order to guarantee the welfare of future generations.**

Our vision, “solar-driven chemistry”, is a long-term innovative scientific and technological endeavour to achieve sustainable chemical production. This ambitious goal will enable the development of the necessary knowledge to accomplish the devices and processes
which progressively will allow the substitution of fossil fuels as the main chemical and energy source. It will also have a large impact on the reduction of greenhouse gas emissions, with huge environmental, societal and economic benefit.

The primary feedstocks for solar-driven chemistry are water (H$_2$O), nitrogen (N$_2$) and carbon dioxide (CO$_2$). The main products of solar-driven chemistry would be molecular hydrogen (H$_2$) obtained through water splitting and a series of carbon-based chemical compounds, obtained through the simultaneous reduction of CO$_2$.

Solar-driven chemistry is a visionary concept, for which many scientific and technical problems still have to be solved. Transfer from basic chemical research to industrial applications usually takes 20 to 30 years. However, intermediate short- and medium-term objectives, which are necessary to enable the long-term goal, can also generate new knowledge, which will provide wider benefits to society and an improvement of the industrial competitiveness.

We wish to point out that this White Paper is not focussed on the conversion of solar energy into electricity (e.g., photovoltaics) or heat (e.g., Concentrating Solar Power (CSP) technologies), but instead targets the use of solar energy directly for the production of fuels, chemicals and materials, thus mimicking nature.
Relevance, Opportunities, Problems, Scenarios

Europe aims to substitute 30% of the fossil carbon used by non-fossil fuel sources by the year 2030. The complete replacement of the current infrastructure of energy production/distribution and petrochemistry would be prohibitively expensive and complicated. Therefore, the only way to guarantee a realistic transition is to integrate alternative raw materials and commodities, not based on fossil carbon but instead on recycled carbon (CO₂), into the existing systems so as to minimize changes of the infrastructure. Reaching this target requires the introduction of “solar-driven chemistry” concepts on a large scale. The produced “solar” fuels and commodities (viz. basic feedstock used in the chemical industry) could be stored, transported and used within existing assets, replacing oil and its refined products.

From the chemical point of view, solar fuels and commodities would be the same as we use today but are derived from water and recycled carbon instead of fossil carbon. In principle, they could be produced everywhere since CO₂, water and solar energy are ubiquitous. The independence from fossil fuels would have a huge geopolitical impact, because it will break down the current situation where few countries have the monopoly of the energy and chemical markets.

In order to accomplish a sustainable renewable energy supply, energy storage is essential. A long-time storage strategy is necessary to balance not only the daily demand cycles, but also seasonal fluctuations. This remains one of the main issues to be solved for renewable energies. Fuels generated from water (hydrogen) or carbon dioxide (carbon monoxide, formic acid, alcohols, hydrocarbons, etc.) using sunlight as the energy source are an extremely attractive green energy storage solution. High energy densities, high volume, and long term and for large storage capacities, solar fuels are a remarkable option, because energy can be stored directly in chemical bonds for an indefinite period of time. Moreover, chemical bonds have an unsurpassed energy storage density. are all attainable. The opportunities for the generation and storage of renewable energy “off-grid"grid” are very important, especially for developing countries, where the economic costs of bringing the grid to the remote areas can be prohibitive. Moreover, energy production and storage systems based on solar fuels is infinitely scalable, from the industrial, through the household, down to the personal level, - making them the ultimate "portable energy".

The scientific and technological challenges to be solved for converting CO₂ into fuels and commodities are still huge and call for an extraordinary effort to mobilize human and economic resources. There are some attempts of large initiatives on solar-driven chemistry-related topics involving integrated academy and industry research at both international and European levels. Notably, major chemical and energy companies forecast that this is a main area for future business. Nevertheless, European efforts need a more effective integration in order to enable a common strategy with clearly defined objectives.
On a laboratory scale it is already possible to produce compounds such as H₂, CO, formic acid, methanol or methane from water, CO₂ and sunlight, to be used as chemicals and fuels. However, there are still many scientific and technological hurdles on the way to the industrial scale-up. Furthermore, the different approaches are sometimes difficult to compare, preventing the identification of the most suitable ones. Benchmarking and standardization are needed. In this context, the Solar Fuels Institute SOFi¹, an international non-governmental initiative, provides, among others, a knowledge map on solar fuels, a tool for solar taxonomy, and a standardization of the information reported in scientific articles. The goal is to implement a consistent database that will generate truly useful knowledge to make results comparable and to identify gaps that need to be filled.

At the present early stage of development, the efficiency of available “solar-driven” processes needs to integrate individual components more effectively into functional devices, which requires a better understanding of several fundamental aspects. Eventually, the implementation of solar-driven chemistry at an industrial scale requires facing some challenges which are illustrated herein, where the focus is on hydrogen and carbon-based chemicals, but other products can be targeted.

Framing the challenges into a time scale, the following scenarios can be outlined.

- In the short term, electricity generated by photovoltaic (PV) devices and transported through the grid could be used for powering commercial water electrolysers that split water into its components and thus generate molecular hydrogen (H₂). This hydrogen can be used directly as a fuel or as an intermediate for the production of carbon-based fuels and chemicals through known thermal routes (Figure 1, centre left). Within the next five years, the realization of the first demonstrators for semi-industrial scale production of H₂ from water and solar energy can be foreseen. Additionally, electricity produced through PV would progressively substitute current electrical technologies, particularly those based on fossil fuels (left box in Figure 1), as already happening in several developed countries. This scenario has, however, some significant weaknesses. The currently best performing water electrolysers based on proton-exchange membranes (PEM) utilize noble metal catalysts, which hinders utilization on the large scale eventually required. The alternative, alkaline electrolysers require steady current-voltage input, which is not compatible with intermittent sources, and would therefore require large increases in grid-based, renewable electricity production with concomitant challenges to balance intermittencies.
- In the short to medium term, the direct production of simple carbon-based compounds of high energy value (e.g., formic acid, formaldehyde, methanol, dimethyl ether, methane, ethanol, acetic acid), avoiding the intermediate production and handling of hydrogen, would be possible by coupling water electrolysis and electrochemical reduction of CO₂ (Figure 1, centre right). Within a decade, it may be expected to realize such processes in prototype demonstrators. However, the disadvantages of coupling such an electrochemical device to a PV system are the same as outlined before.
- In the medium to long term, hydrogen (Figure 1, left) or carbon-based compounds
Solar-driven chemistry (Figure 1, right) could be produced directly by photoinduced reactions. Instead of generating electrons through PV and using them in electrochemical cells, the solar energy would be captured and used in an integrated system, such as photoelectrochemical cells (PECs) or photocatalytic devices (see below) that are developed to function in intermittent sunlight. The realization of these devices will require at least 15–20 years. Large-scale direct H₂ production in water-splitting systems powered by sunlight might be feasible on a shorter time scale than producing carbon-based fuels and commodities (see below), because proton reduction to hydrogen gas is far less difficult than CO₂ reduction.

![Diagram of solar-driven chemical processes](image)

**Figure 1** Options for solar-driven chemical processes.²

The realization of this timeline will largely depend on the intensity of the resources dedicated to research and development, but also on the possibility of realizing efficient instruments to coordinate and intensify the research in this sector. Lab-scale technological demonstrators (Technological Readiness Levels 3-4) have already been developed for photoelectrochemical H₂ evolution and CO₂ reduction, but several years are still necessary to make them efficient, safe, robust and economically viable.
Targets and Approaches

Hydrogen

Hydrogen has sparked wide interest because its photochemical production from water and use is not coupled with the production of CO\(_2\). There are serious technical drawbacks, however, associated with its storage and transport. Hydrogen produced from water can be used for the production of hydrocarbons (which are easily stored and transported) or chemical commodities by means of established routes. In particular, syngas – synthesis gas, a mixture containing varying proportions of CO and H\(_2\) – is the most interesting option and already has high industrial relevance. This mixture can be converted into hydrocarbons or alcohols through the established Fischer-Tropsch process.

Current production methods of hydrogen are primarily based on fossil fuels and include steam reforming of natural gas or liquid hydrocarbons and the gasification of coal. The main non-fossil carbon-based source of hydrogen is water. However, the "water-splitting" process mentioned above is very energy-demanding and solar energy cannot be used directly, because water does not extensively absorb sunlight. At present, photovoltaic panels coupled with electrolysers with the aid of catalysts is the most viable route to water splitting (with the disadvantages mentioned above). CO\(_2\) can be dissociated into CO and oxygen using the same technologies but with different catalysts. In a nutshell, it is possible to use solar energy, water and CO\(_2\) to produce syngas and thus liquid fuels. However, further scientific and technical advancements are needed, particularly in the development of direct routes, which do not encompass the use of electrolysers. Current electrolysers have problems with large upscaling and/or handling intermittent electricity inputs, as stated above, and run with typically 30% energy losses, from electrical to chemical energy. A direct process to solar fuels may, in principle, avoid the inevitable losses of an intermediate conversion step and lead to lower balance-of-systems costs for a single device, compared to separate photovoltaics plus electrolysers.

In electrolytic processes, light harvesting and catalysis are well separated, while they are intimately coupled in “direct” photochemical or photoelectrochemical processes. In such processes, absorption of solar radiation and charge separation, i.e. creation of electrons and holes ("light harvesting"), is achieved by means of a photoactive material interacting with catalysts which use electrons to produce the fuel, or holes to generate oxygen and protons from water. The photoactive material can be a molecular compound (synthetic unit or a natural photosystem isolated from plants or algae) or an inorganic semiconductor. Similarly, the catalyst can be a molecular species (an enzyme or a synthetic compound) or an inorganic solid (for example a metal or metal oxide). There are several strategies for combining such photoactive systems with catalysts to generate photocatalysts for oxygen and hydrogen production.

The key components needed for realizing a homogeneous system for water splitting – as inspired by natural photosynthesis – are schematically outlined in Figure 2. It comprises
In such a system, sunlight triggers a flow of electrons from the water oxidation catalyst (which extracts 4 electrons and 4 protons from two water molecules, releasing a molecule of oxygen) to the \( \text{H}_2 \) evolution catalyst. Electron transfer occurs directly at the interfaces between the different components. The challenge for chemists is to organize the different constituents and foster the electron flow.

A related concept is a system, where a suspension of semiconductor nanoparticles is used instead of molecular dyes and catalysts. The nanoparticles are made from different materials for light-harvesting, which are decorated with catalyst particles of a different material.

An alternative (and presently more developed) approach is photoelectrochemical cells (PEC), with solid electrodes. These usually consist of:
- a semiconductor p/n junction material that absorbs visible light and separates electrons and holes,
- a catalyst material or nanoparticles deposited on the semiconductor for oxygen production,
- a catalyst material or nanoparticles for hydrogen production,

More recently, hybrid systems have been developed which utilize both solid materials and
molecules, performing different parts of the processes. Typically, the catalysts are molecule-based, and sometimes also the photoactive system, while charge separation and charge transport is carried out by a semiconductor material. In one approach, the molecular constituents are immobilized on electrodes and these electrodes are integrated into a PEC. A possible architecture of a tandem PEC (i.e. a PEC with two photoactive electrodes) is depicted in Figure 3. The main components of such a PEC are:

- a photoanode to be exposed to sunlight which is functionalized with a photosensitizer (PS) in turn linked to a photocatalyst able to oxidize water,
- a proton-permeable membrane that enables fast transport of protons between the electrode compartments and the separation of the two generated gases,
- a photocathode with a similar structure as the photoanode (i.e., with a photosensitizer and a catalyst) and able to convert protons into hydrogen.

The electrodes are connected with a wire to allow the electron flow between the two redox sites as mentioned above. It is also possible to use a single photo-active electrode in the same arrangement, with a “dark” counterpart that consists only of the conducting material and a catalyst.

![Figure 3](image)

Figure 3  Schematic representation of a tandem photoelectrochemical cell (PEC) for water splitting (see text for explanation). (WOC = water oxidation catalyst; PRC = proton (H+) reduction catalyst, hν = solar radiation).

Challenges for Research

- Improving light harvesting, understanding interfacial energy/electron transfer processes and finding means to prevent charge recombination
- Developing cheaper and more stable electro- and photo-catalysts
Carbon-based Compounds
High-energy processes: production of reduced forms of CO₂.

CO₂ is a cheap and abundant molecule, however only a few industrial processes use it at present, for example in the production of carboxylic acids, organic carbonates, urea or methanol. Such processes utilize about 180 Mt/y of CO₂, to be compared with over 36 Gt produced annually in the world by human activities and released in the atmosphere. CO₂ is an extremely stable molecule, which requires a lot of energy for its conversion to useful energy-rich chemicals. Furthermore, it is kinetically inert because the reduction processes require high activation energy. Transforming CO₂ is therefore a scientifically challenging problem, requiring, *inter alia*, novel catalysts.

One possibility is “CO₂ hydrogenation”, *i.e.*, the catalytic reaction of CO₂ with hydrogen, which is an established process. In a first step towards solar-driven chemistry, hydrogen produced by conventional methods could be replaced by hydrogen obtained from water through photovoltaic-based electrolysis (see above). CO₂ hydrogenation is attractive because it can transform CO₂ and hydrogen directly into gaseous or liquid compounds, such as formic acid, methanol, methane and larger hydrocarbon molecules, which are much more easy to handle and transport than hydrogen itself and hence more appealing for industrial production with existing infrastructures. Both homogeneous and heterogeneous catalysts can be used, including some based on cheap metals (Fe, Cu, Ni, Mn).

The most innovative and appealing strategy for the exploitation of CO₂ would be direct use of solar energy. Conceptually, the idea is to mimic at the industrial level what happens in green plants, green algae and cyanobacteria that adsorb sunlight and convert CO₂ and water into complex high-energy molecules through photosynthetic systems, with oxygen as a “by-product”. The process involves two multi-step reactions: the oxidation of water to oxygen (similar to what was described above for photo- and electrochemical hydrogen production) and the selective reduction of CO₂. In plants, CO₂ is reduced in the so-called Calvin cycle using NADH generated from electrons released in the photochemical water oxidation step (Figure 4).
Although CO$_2$ reduction in natural systems is not based on CO$_2$ electroreduction or hydrogenation, some key features may serve as a blueprint for solar-driven synthesis of carbon compounds. From nature we only need to mimic the key principles, the utilization of energy from sunlight to oxidise water and reduce CO$_2$ into a chemical fuel. In addition, we should aim to be more efficient than the natural processes (natural photosynthetic organisms have a net light-to-chemical-energy conversion of 4-5% at best, in general much lower than that. Microalgae may reach 6-8% in nature and 8-12% in photobioreactors). 

As for hydrogen, there are two main strategies to accomplish photochemical CO$_2$ reduction:

- Homogeneous light-driven reduction accomplished through molecular compounds that are used as photosensitizers and/or catalysts. The photosensitizer compounds should be able to absorb a significant proportion of the solar spectrum, have long-lived excited states and be sufficiently stable. The catalysts are typically transition metal complexes, which couple light-induced charge separation to the catalytic fuel-forming and water oxidation reactions.

- Photoelectrochemical reduction using electrodes that are suitably functionalized with catalysts. As for hydrogen production, hybrid systems are emerging with molecular catalysts, and also dyes on semiconductor electrodes.

Photocatalytic systems must be optimized to improve both light absorption and catalytic capability, which is accomplished by ligand design and variation of the metal centre. The catalyst must orchestrate the coupled transfer of multiple electrons and protons, and the conversion of the CO$_2$ substrate. Molecular catalysts are considered especially suitable, because they allow, in principle, a better control of bond-breaking and -making processes in small molecules. Following the concepts provided by nature, an optimized design is possible based on both a deep mechanistic understanding of the active site of enzymes and of the role
of the matrix surrounding the catalytic centre, for example in capturing substrates, channelling electrons and releasing products. During charge separation, the intermediary acceptor/catalytic site needs sufficient stability in the reduced state, which often depends on the set of ligands surrounding a metal centre. Currently most photocatalysts reported for CO$_2$ reduction are based on Ru and Re, but systems based on more abundant first row transition metals are emerging.

In photobioelectrochemical systems (PBES), bacteria interact with solid electrodes by exchanging electrons with them, either directly or via redox mediators. A possible application is the electrochemical reduction of carbon dioxide to methane.

**Challenges for Research**

- Better understanding of the complex molecular mechanisms and key steps in natural and artificial photosynthesis (e.g., C-C bond formation, multiple electron/proton transfer)
- Developing cheaper, more selective and more stable (photo-)catalysts for CO$_2$ reduction
- Finding cheap solutions to concentrate and use CO$_2$ from the atmosphere

**Other Chemicals**

*Ammonia* is one of the most energy-intensive products of the chemical industry. Its synthesis by solar-driven routes from air and water at ambient temperature and pressure is a grand challenge. Presently, around 1-2% of the world’s energy is used for industrial nitrogen fixation in the form of ammonia and derive products, which are at the basis of fertilizers. Ammonia is currently produced from nitrogen (separated from air) and hydrogen (obtained from natural gas) under severe conditions. However, some organisms can synthesise ammonia from air and water at ambient temperature and pressure. In an attempt to mimic these processes, metallo-enzymes of natural nitrogenases have been studied for over forty years. The key issue for nitrogen fixation is understanding the redox processes at the catalytic metal centers. Novel synthetic electrocatalysts for N$_2$ to ammonia conversion have been recently reported and some electrochemical syntheses of organic amines have been achieved, though with still non-exploitable turnover numbers. Such systems are proof-of-concept, but the use of solar energy to replace fossil carbon-based electricity may open radically new production perspectives. Producing ammonia on an industrial scale using nitrogen, water, and photo-driven catalysts, would save large amounts of natural gas and energy. In addition, ammonia (and then fertilizers) could be produced on a distributed scale, avoiding the need of large plants (as currently) with high local environmental impact and the costs and risks associated with the transport of large volumes of chemicals.

*Hydrogen peroxide* (H$_2$O$_2$) is a very clean oxidant, because it produces water as the reaction by-product. However, its use is still limited at the industrial scale because of the costs of production, high energy intensity and environmental issues. To make the process economic, large-scale plants for H$_2$O$_2$ industrial production are necessary. H$_2$O$_2$ may find use also in...
various environmental cleaning applications, from waste water treatment to soil remediation, when produced at lower cost and with greener technologies. \( \text{H}_2\text{O}_2 \) may be potentially produced in an electrochemical cell using solar energy, water and \( \text{O}_2 \) as reactants, possibly enabling small scale productions and avoiding the safety issues related to transport.

There is a whole range of other inorganic and carbon-based chemicals which presently require energy-intensive production processes and which could be produced in the future using solar-driven chemical processes. The solar-driven approach will not only allow the use of renewable energy sources instead of fossil fuels, but will also foster a shift to a new model of distributed production which avoids large concentration of plants, with the related impact on the environment. It is therefore a new model of industrial chemical production which may largely benefit the industrial competitiveness.

**Challenges for Research**
- Develop electro- and photocatalysts, as well as electrodes for ammonia production
- Develop new catalysts for the synthesis of \( \text{H}_2\text{O}_2 \), driven by renewable energy
Materials Constraints

One of the most important aspects – besides device design, efficiency, durability and scalability – is the availability of materials to manufacture the infrastructure that is needed. A key issue is that the possibility of making fuels and chemicals with solar energy is not limited by the number of photons reaching the Earth, but may be subject to limitations with regard to the materials needed to manufacture light-converting devices, catalysts, electrodes, energy accumulators, etc. The required materials obviously must come from the Earth’s crust, which is a large but ultimately limited reservoir.

As outlined above, the feasibility of solar-driven chemistry has been demonstrated on the laboratory scale, but most of the laboratory-scale devices made so far use chemical elements which are scarcely available on the Earth crust (e.g., platinum group or some rare earths elements). Unfortunately, less abundant elements are currently still essential for renewable and efficient energy technologies and it is not yet clear whether such elements are available to a sufficient extent for enabling solar-driven chemistry on a large scale. This can be a formidable obstacle for application on a larger scale and future market penetration. Furthermore, a few countries control the majority of rare mineral commodities, rising also geopolitical issues.

An example of the so-called material trap is the development of a solar hydrogen generator by the National Renewable Energy Laboratory in Colorado. This system was a breakthrough as it was four times as efficient as natural photosynthesis. It was based, however, on rare elements, including platinum electrodes and gallium-indium-based photovoltaic cells. Insufficient stability was an additional problem. Although this approach proved that such systems can be successfully developed, its cost remains prohibitive for large-scale commercial applications primarily due to materials limitations.

The materials to be used in industrial processes based on solar-driven chemistry should have low environmental impact throughout the entire life cycle, and be stable for many years under exposure to sunlight. Replacement of some of the most expensive, less abundant and potentially harmful materials that are currently being tested and used with cheaper, more abundant, non-toxic materials is therefore essential for the implementation of a solar-driven chemistry. At present, however, the majority of metals have virtually no substitutes for most applications, hence the development of methods for the recovery of less abundant elements (“urban mining”) is another urgent need for implementing a sustainable path to solar-driven chemistry.

Challenges for Research

- Replacement of critical elements with less expensive and more abundant ones.
- Development of industrially-feasible recovery methods for less abundant elements.
Conclusions

Solar-driven chemistry is a radical paradigm shift in chemical production, which will have a high impact on the competitiveness and sustainability of the European industrial system. It has the potential to contribute significantly to a fossil-independent supply of feedstock for the chemical industry and to green fuels for all applications.

Solar-driven chemistry can create knowledge-driven competitiveness for Europe’s industrial production, while preserving jobs and the environment. In order to accomplish this ambitious goal, a broad and inclusive action driven by chemists is needed. It requires a large integrated and synergic approach to meet these ambitious objectives, not least research that encompasses catalysis, electrochemistry, photochemistry, nanosciences, in concert with semiconductor physics, engineering, biosciences and social sciences.

Many fundamental questions and problems related to transferring laboratory results to industrial applications still have to be solved. Implementation of solar-driven chemistry is a big challenge, with high impact for future generations, not only in science, industry and economy, but also within society as a whole. It will take several decades to reach all the ambitious goals outlined in this document, but short-term and intermediate results will undoubtedly generate huge benefits. Clearly, realising solar-driven chemical technologies requires a strong, concerted and unconventional support to basic and applied research.

References

1. www.solar-fuels.org
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