

SUN-to-LIQUID

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Project Summary

The SUN-to-LIQUID project (<https://www.youtube.com/watch?v=laUe23OhHXg>) developed the technology to produce renewable drop-in fuels that have the potential to minimize the carbon footprint of the aviation sector at the global scale. It uses abundant feedstock water, CO₂ and solar energy without conflict with arable land. The core conversion technology is a thermochemical redox cycle driven by concentrated solar energy, utilizing the entire solar spectrum and operating at thermodynamically efficient high temperatures, to co-split water and CO₂ into high-quality syngas that is further processed to clean hydrocarbon fuels. Following laboratory-scale experiments, the SUN-to-LIQUID project advanced this technology with approximately a 10-fold increase of solar radiative power and a 3-fold improvement in solar-to-syngas energy efficiency. Solar fuel plants are expected to provide local jobs in rural areas and increased energy security, as already proved with concentrated solar power plants. The innovation has been demonstrated at technology readiness level (TRL) 5 on-sun in an integrated solar tower chemical plant. Its characteristics both of the core innovation as well as the system-level performance and climate mitigation potential, techno- and socio-economic impact have been published according to scientific standards.

Project Motivation and Objective

The motivation of the SUN-to-LIQUID project was to develop the scientific and technological basis for the efficient and economical production of aviation fuel directly from water, CO₂ and solar energy, with greatest replication potential for the development of economically challenged regions with arid climate, high levels of solar radiation and non-arable land. Two particularly important challenges and targets in the global agenda are: First, the Paris Agreement, concerning the limitation of global warming to 1.5°C, which according to the IPCC 2018 Special Report requires the implementation of renewable energy and technologies for deep de-carbonization. Second, the aviation industry has set a target of 50% reduction of CO₂ emission in 2050 compared to 2005 and complete global de-carbonization by 2060 (ATAG target). Those ambitious targets of the aviation sector are also a driver for the future development of technologies to produce drop-in-capable renewable aviation fuel in the order of 500 Mt/yr by 2050. While rail, road, or maritime transport may utilize electricity or renewable hydrogen for propulsion, aviation is expected to rely on high-energy-dense liquid hydrocarbon fuels for many decades to come. Biofuel technology faces severe challenges to meet sustainability and availability requirements at the scale of future global fuel demand. The advent of synthetic solar jet fuels offers a truly sustainable and scalable perspective as the future global fuel demand can be met by utilizing less than 1% of the global arid land. Thus there is no land competition with food or feed production. The SUN-to-LIQUID project was based on a scientific breakthrough in redox chemistry (*Energy & Env. Science* 10, 1142-1149, 2017), and the favorable techno-economic prospects of a solar-driven fuel production process for the global supply of clean liquid fuels (*Sustainable Energy & Fuels* 4, 3992, 2020). The main objective was to demonstrate the feasibility and scalability of the entire production path to sustainable synthetic hydrocarbon fuel and to establish a robust roadmap to commercial solar fuel production. The specific objective was to demonstrate solar fuel production with a record solar-to-syngas energy conversion efficiency in an integrated plant using an advanced high-flux ultra-modular solar heliostat field, a 50-kW solar reactor, and optimized redox materials.

Project Innovation and Results

The SUN-to-LIQUID project encompasses basic and applied R&D of the solar thermochemical fuel path via material research, solar reactor development, development of a high-flux concentrating modular solar field and tower infrastructure, computational simulation, and process optimization. Research and development for the solar reactor technology included fundamental thermodynamic and kinetic analyses of the redox active material ceria, development of advanced porous ceramic structure with superior heat and mass transport properties, and the fabrication, testing, modelling, and optimization of a 50-kW solar thermochemical reactor. Research and development related to the high-flux solar concentrator, consisting of a sun-tracking heliostat field, included novel fabrication methods for the focusing mirrors, new field and focus characterization methods and advanced control systems for stable delivery of flux intensities beyond 2000 suns at the reactor's aperture, exceeding the solar concentration ratios typically obtained in CSP plants for electricity generation. The 50-kW solar reactor achieved a new record value of the solar-to-fuel energy conversion efficiency of 5.6% for pure CO₂-splitting and 4.1% for co-splitting of CO₂ and H₂O, and a pioneering long-term routine operation on a daily basis of > 100 cycles. The synthesis gas was further processed on-site via Fischer-Tropsch to a mixture of kerosene, diesel, and naphtha. Thus the project contributed to performance/cost ratio increase and improved fabrication technology both for the solar reactor and the heliostat field (including cost-effective fabrication by compliant mirror beam shaping). Better component lifetime was demonstrated through the multiple-cycle production campaign. The technological research tasks were accompanied by system-level techno-economic, environmental and social impact analyses of a future commercial plant based on the demonstrated fuel production pathway in order to quantify the future improved environmental profile of this fuel production innovation. The life cycle assessment shows GHG emission relative to conventional fossil jet fuel reduced by over 80%, with emissions approaching zero when construction materials (e.g. steel, glass) are manufactured using renewable energy. The very low water demand of 3-5 l per liter of liquid fuel produced is much smaller compared to 2nd-generation biofuels. All results were published according to scientific standards in several open-access peer-reviewed journals. The non-CO₂ climate impact specific to aviation, e.g. by contrail formation, is expected to be significantly reduced because of lower particulate matter emission of the solar fuel, which is sulfur-free and contains very low levels of aromatic content, resulting also in improved air quality and reduced health risks around airports. The long-term goal is solar fuel production at a global scale, which will require replication of solar fuel plants distributed globally, promoting economic benefits and enhancing supply security.

Role of the Applicants

This proposal is submitted by the SUN-to-LIQUID consortium, which executed the complete EU-project. The applicants, acting on behalf of the consortium, and their roles are: ETH Zurich (Switzerland) – solar reactor design, fabrication and operation; IMDEA Energy (Spain) – high-flux solar tower facility design, implementation and operation; Bauhaus Luftfahrt e.V. (Germany) – project coordination, system-level analysis of environmental, techno- and socio-economic impact. Further consortium members DLR – German Aerospace Center (Germany), HyGear Technology and Services B.V. (the Netherlands), Abengoa Research SL (Spain), and ARTTIC Innovation GmbH (Germany).

Annex – Links to Supporting Innovation, References and Photos and Project Documentation

Links: Project website <https://www.sun-to-liquid.eu/>; EU-website <https://cordis.europa.eu/project/id/654408/results>.

Annex A – Photos and visualization of results; **Annex B** – List of selected peer-reviewed scientific publications.

Annex A -Photos and Visualization of Results

The image gallery below illustrates key elements of the SUN-to-LIQUID project:

Fig. 1: Scheme of the solar fuel processing plant to CO₂-neutral hydrocarbon fuels using sunlight.

Fig. 2: Schematic of the 2-step thermochemical redox cycle for splitting CO₂ and H₂O to produce syngas.

Fig. 3: The solar chemical reactor technology

Fig. 4: Representative solar redox cycle and day solar run for co-splitting H₂O and CO₂.

Fig. 5: The integrated solar-thermochemical plant in Madrid, Spain

Fig. 6: The result of the life-cycle analysis of net carbon intensity for renewable jet fuel from a future commercial plant.

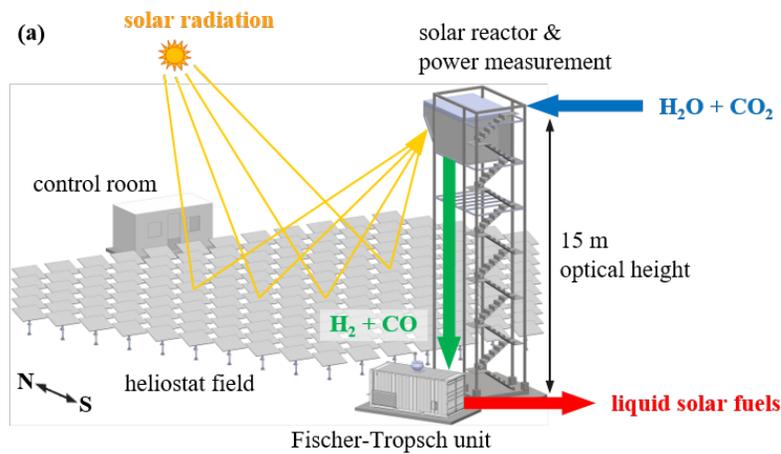


Figure 1. Scheme of the solar fuel processing plant to CO₂-neutral hydrocarbon fuels using sunlight. The SUN-to-LIQUID project serially integrates three energy conversion units: 1) the collection and concentration of solar energy using a solar tower configuration; 2) the solar chemical reactor which converts CO₂ and H₂O to a desired mixture of CO and H₂ (syngas); 3) the Fischer-Tropsch unit which converts syngas to liquid hydrocarbons. These are carbon-neutral synthetic fuels because they release only as much CO₂ during their combustion as was previously utilized for their production.

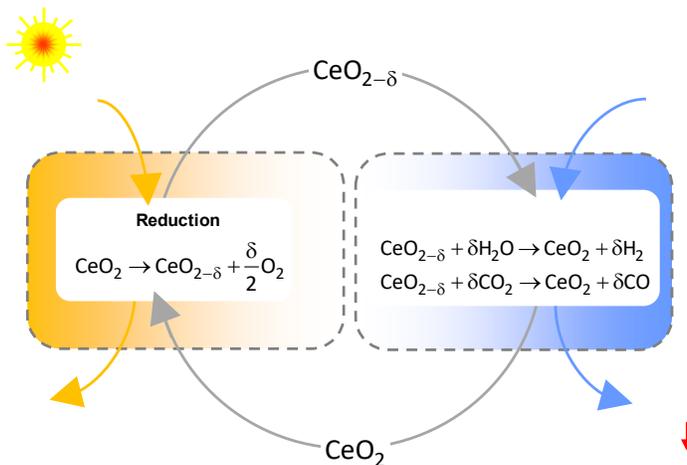


Figure 2. Schematic of the 2-step thermochemical cycle for splitting CO₂/H₂O into separate streams of CO/H₂ and O₂ via reduction-oxidation (redox) reactions using non-stoichiometric ceria (CeO₂). In the first endothermic step, ceria is thermally reduced to generate O₂ using concentrated solar process heat. In the second exothermic step, the reduced ceria is re-oxidized with CO₂ and H₂O to generate CO and H₂, respectively. δ denotes the nonstoichiometry – the measure of the redox extent and thereby the fuel yield per cycle.

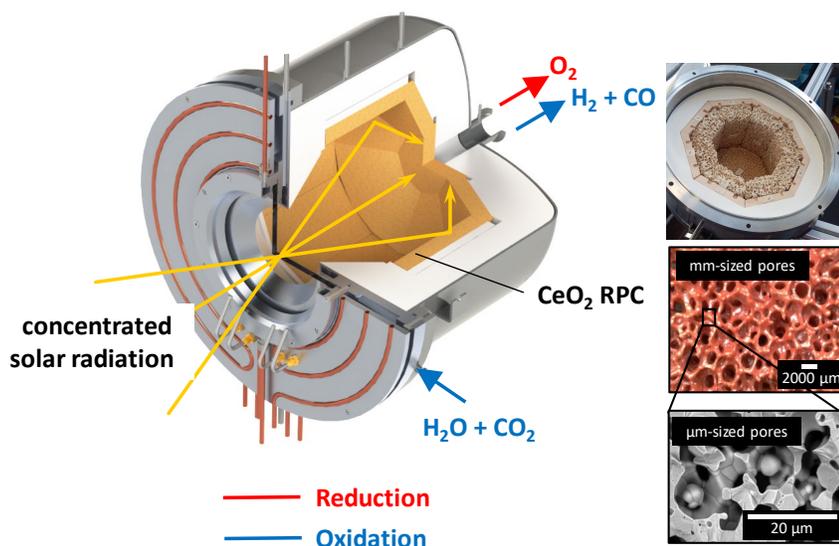


Figure 3. Schematic of the solar reactor configuration for splitting CO₂ and H₂O. It comprises a cavity-receiver containing a reticulated porous ceramic (RPC) structure made of ceria directly exposed to high-flux solar irradiation. Red arrow: endothermic reduction generating O₂ is performed using concentrated solar energy. Blue arrow: exothermic oxidation with CO₂ and H₂O generating CO and H₂ (syngas). Photographs of the solar reactor, showing the front face of the solar reactor and its interior containing the ceria RPC structure with dual-scale porosities in the mm and μm ranges for enhanced heat and mass transport.

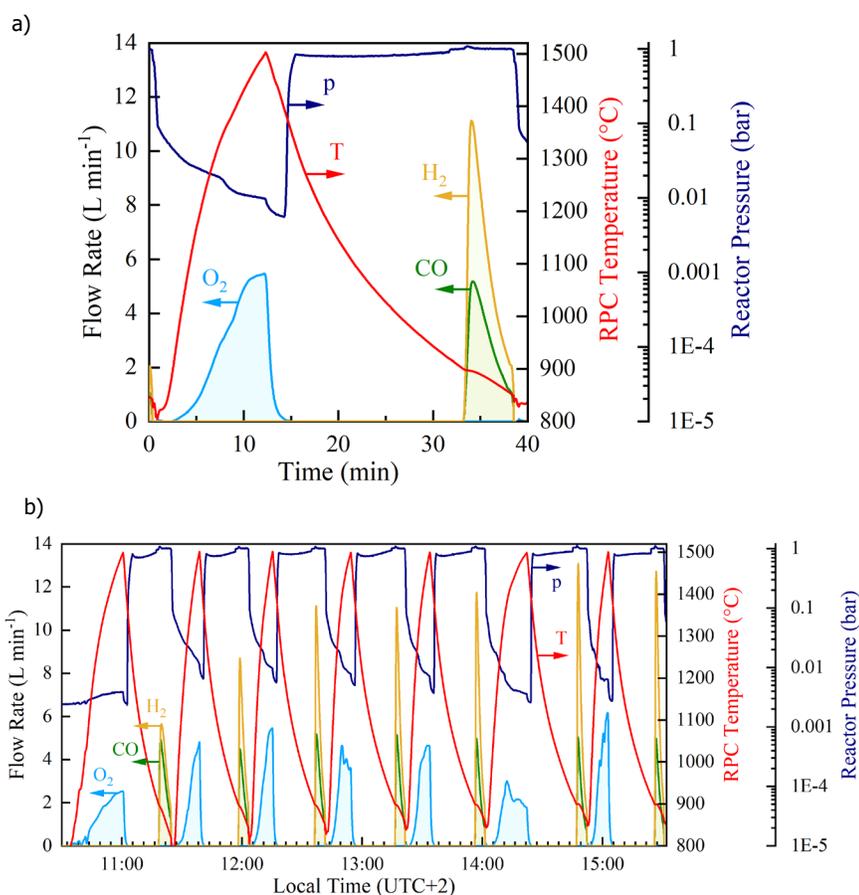


Figure 4: a) Representative solar redox cycle for co-splitting H₂O and CO₂. Temporal variation of the nominal temperature and absolute pressure of the solar reactor, and the product gas composition during the reduction and oxidation steps. The operational conditions are: total feeding rate of H₂O and CO₂ of 81 L/min, oxidation starting temperature of 900 °C, and feeding molar ratio H₂O:CO₂ of 5.5. L denotes standard litre, mass flow rates calculated at 273 K and 1 atm. b) Representative day solar run with 7 consecutive redox cycles for co-splitting H₂O and CO₂. Temporal variation of the reactor nominal temperature, reactor absolute pressure, and product gas composition during the reduction and oxidation steps. The operational conditions are: total feeding rate of H₂O and CO₂ of 81 L/min, oxidation starting temperature of 900 °C, and feeding molar ratio H₂O:CO₂ of 5.5. L denotes standard litre, mass flow rates calculated at 273 K and 1 atm.



Figure 5. SUN-to-LIQUID Integrated plant in operation including solar heliostat field, thermochemical reactor (on top of tower), and Fischer-Tropsch conversion (in container to the right). (Image by Abel Valdenebro ©ARTTIC 2019).

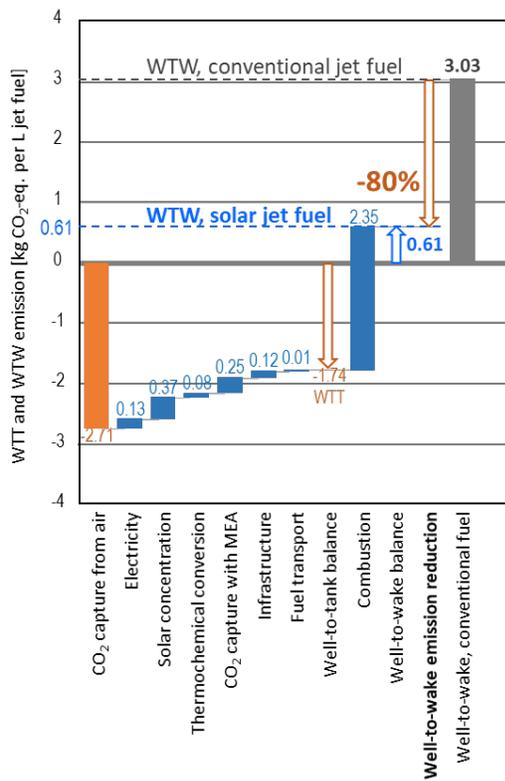


Figure 6. Net greenhouse gas emission from the production and combustion of synthetic solar jet fuel (0.61 kg CO₂-equivalent GHG per litre) is 80% less than conventional jet fuel, which emits 3.0 kg CO₂-equivalent GHG per litre (adapted from: Falter et al., Sustainable Energy and Fuels, 4, 2020)

Annex B – List of selected peer-reviewed scientific publications

- 1 **A solar tower fuel plant for the thermochemical production of kerosene from H₂O and CO₂**
Zoller S., Koepf E., Nizamian D., Stephan M., Patané A., Haueter P., Romero M., Gonzalez-Aguilar J., Lieftink D., de Wit E., Brendelberger S., Sizmann A., Steinfeld A.,
Joule 6, pp. 1606-1616 (2022);
DOI: 10.1016/j.joule.2022.06.012
- 2 **An integrated techno-economic, environmental and social assessment of the solar thermochemical fuel pathway**
Falter C., Valente A., Habersetzer A., Iribarren D., Dufour J.,
Sustainable Energy & Fuels 4, 3992 (2020);
DOI: 10.1039/D0SE00179A
- 3 **Geographical Potential of Solar Thermochemical Jet Fuel Production**
Falter C., Scharfenberg N., Habersetzer A.,
Energies 13, 802 (2020);
DOI: 10.3390/en13040802
- 4 **A 50 kW solar thermochemical reactor for syngas production utilizing porous ceria structures**
Zoller S.,
Diss. ETH No. 26451 (2020);
DOI: 10.3929/ethz-b-000426499
- 5 **Solar-Driven Thermochemical Production of Sustainable Liquid Fuels from H₂O and CO₂ in a Heliostat Field**
Romero R., González-Aguilar J., Sizmann A., Batteiger V., Falter C., Steinfeld A., Zoller S., Brendelberger S., Lieftink D.,
Proc. ISES-SWC2019 Solar World Congress, Santiago, Chile, Nov. 4-7, 2019;
DOI: 10.18086/swc.2019.23.02
- 6 **Heat transfer model of a 50 kW solar receiver-reactor for thermochemical redox cycling using cerium dioxide**
Zoller S., Koepf E., Roos P., Steinfeld A.,
Journal of Solar Energy Engineering, Vol. 141, 021014-11 (2019);
DOI: 10.1115/1.4042059
- 7 **Liquid Fuels from Concentrated Sunlight: An Overview on Development and Integration of a 50 kW Solar Thermochemical Reactor and High Concentration Solar Field for the SUN-to-LIQUID Project**
Koepf E., Zoller S., Luque S., González-Aguilar J., Romero M., Steinfeld A.,
SolarPACES Conference 2018, Casablanca, Morocco, Oct. 2-5, 2018; *AIP Conference Proceedings* 2126, 180012 (2019);
DOI: 10.1063/1.5117692
- 8 **Water Footprint and Land Requirement of Solar Thermochemical Jet-Fuel Production**
Falter C., Pitz-Paal R.,
Environmental Science & Technology 51 (21), 12938-12947 (2017);
DOI: 10.1021/acs.est.7b02633

More: visit <https://www.sun-to-liquid.eu/page/en/scientific-publications.php>

Publication on details of the innovation

“A solar tower fuel plant for the thermochemical production of kerosene from H₂O and CO₂”,

Zoller S., Koepf E., Nizamian D., Stephan M., Patané A., Haueter P., Romero M., Gonzalez-Aguilar J., Lieftink D., de Wit E., Brendelberger S., Sizmann A., Steinfeld A.,

Joule, Vol. 6, pp. 1606-1616, 2022.

<https://doi.org/10.1016/j.joule.2022.06.012>

Abstract

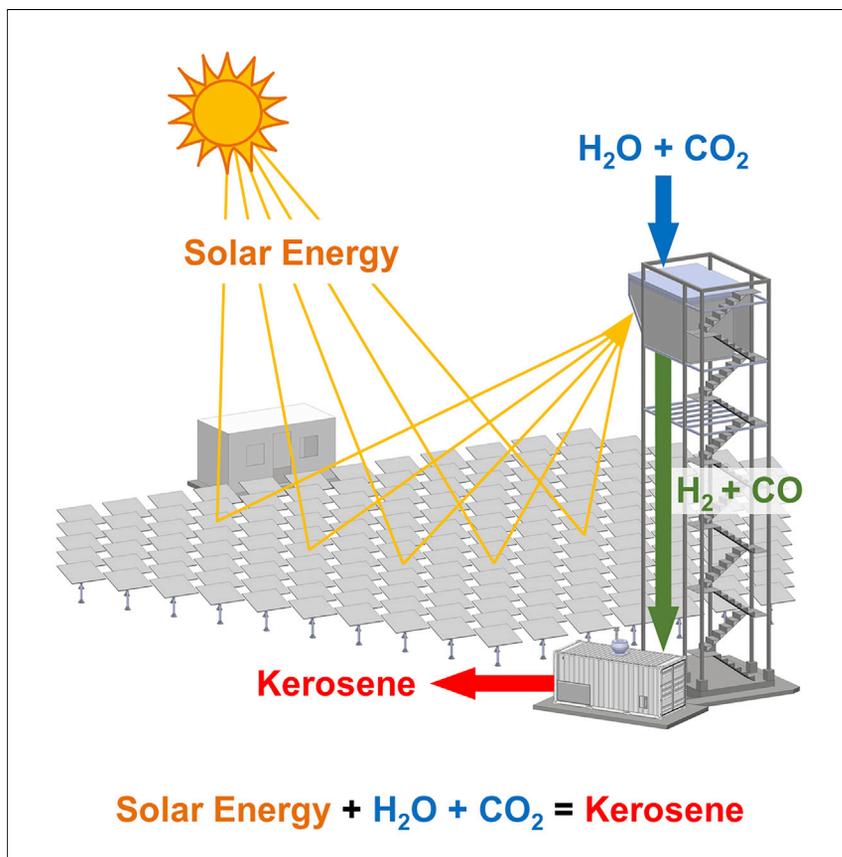
Developing solar technologies for producing carbon-neutral aviation fuels has become a global energy challenge, but their readiness level has largely been limited to laboratory-scale studies. Here we report on the experimental demonstration of a fully-integrated thermochemical production chain from H₂O and CO₂ to kerosene using concentrated solar energy in a solar tower configuration. The co-splitting of H₂O and CO₂ was performed via a ceria-based thermochemical redox cycle to produce a tailored mixture of H₂ and CO (syngas) with full selectivity, which was further processed to kerosene. The 50 kW solar reactor consisted of a cavity-receiver containing a reticulated porous structure directly exposed to a mean solar flux concentration of 2,500 suns. A solar-to-syngas energy conversion efficiency of 4.1% was achieved without applying heat recovery. This solar tower fuel plant was operated with a setup relevant to industrial implementation, setting a technological milestone towards the production of sustainable aviation fuels.

Context, Scale, and Significance:

The aviation sector, which strongly relies on fossil-derived kerosene, is responsible for vast amounts of anthropogenic greenhouse gas emissions. To avoid these emissions, solar energy can be leveraged to efficiently produce sustainable drop-in fuels (e.g. kerosene). Solar-made kerosene can replace fossil-derived kerosene and further make use of the existing global jet fuel infrastructures for its storage, distribution, and end-use in jet engines. These infrastructures are particularly critical for the long-haul aviation sector.

For the first time, the thermochemical production of kerosene using solar energy, water, and CO₂ is demonstrated in a fully-integrated solar tower fuel plant. This work, realized within the framework of the EU Horizon 2020 project SUN-to-LIQUID, advances the technological readiness level of solar fuels production by demonstrating the technical feasibility of the entire sun-to-liquid fuel process chain. We have evaluated the performance of the solar reactor – the cornerstone technology – based on five primary metrics (namely: reaction selectivity, syngas quality, fuel purity, energy efficiency, and material stability), and experimentally validated its stable operation and full integration in the solar tower fuel plant. This pioneer technological demonstration, performed at a pilot scale relevant to industrial implementation, represents a critical milestone on the path towards the production of sustainable aviation fuels.

Article

A solar tower fuel plant for the thermochemical production of kerosene from H_2O and CO_2 

For the first time, the thermochemical production of kerosene using solar energy, water, and CO_2 is demonstrated in a fully integrated solar tower fuel plant. Solar-made kerosene can replace fossil-derived kerosene and further make use of the existing global jet fuel infrastructures and engines, which are particularly critical for the long-haul aviation sector. This pioneer technological demonstration, performed at a pilot scale relevant to industrial implementation, represents a critical milestone on the path toward the production of sustainable aviation fuels.

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Highlights

Entire process chain from H_2O and CO_2 to solar kerosene realized in a solar tower

50-kW solar reactor demonstrated for ceria-based thermochemical redox splitting

Consecutive redox cycling produced syngas suitable for FT synthesis

4.1% solar-to-syngas energy efficiency achieved without implementing heat recovery

Article

A solar tower fuel plant for the thermochemical production of kerosene from H₂O and CO₂

Stefan Zoller,¹ Erik Koepf,^{1,*} Dustin Nizamian,¹ Marco Stephan,¹ Adriano Patané,¹ Philipp Haueter,¹ Manuel Romero,² José González-Aguilar,² Dick Liefstink,³ Ellart de Wit,³ Stefan Brendelberger,⁴ Andreas Sizmann,⁵ and Aldo Steinfeld^{1,6,*}

SUMMARY

Developing solar technologies for producing carbon-neutral aviation fuels has become a global energy challenge, but their readiness level has largely been limited to laboratory-scale studies. Here, we report on the experimental demonstration of a fully integrated thermochemical production chain from H₂O and CO₂ to kerosene using concentrated solar energy in a solar tower configuration. The co-splitting of H₂O and CO₂ was performed via a ceria-based thermochemical redox cycle to produce a tailored mixture of H₂ and CO (syngas) with full selectivity, which was further processed to kerosene. The 50-kW solar reactor consisted of a cavity-receiver containing a reticulated porous structure directly exposed to a mean solar flux concentration of 2,500 suns. A solar-to-syngas energy conversion efficiency of 4.1% was achieved without applying heat recovery. This solar tower fuel plant was operated with a setup relevant to industrial implementation, setting a technological milestone toward the production of sustainable aviation fuels.

INTRODUCTION

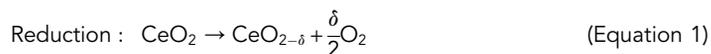
For the foreseeable future, kerosene will be indispensable as a jet fuel for long-haul aviation due to its high specific gravimetric energy density and compatibility with the existing global fuel infrastructure. However, approximately 5% of current anthropogenic emissions causing climate change are attributed to global aviation, and this number is expected to increase.¹ An alternative to conventional kerosene derived from petroleum is kerosene synthesized from syngas—a specific mixture of H₂ and CO—via the established Fischer-Tropsch (FT) synthesis process. The technological challenge, however, is to produce renewable syngas from H₂O and CO₂ using solar energy. The solar-driven thermochemical splitting of H₂O and CO₂ via a two-step metal oxide redox cycle can meet this challenge.² Such a process offers a thermodynamically favorable pathway to syngas production because it uses the entire solar spectrum as the source of high-temperature process heat for effecting the thermochemical conversion, and it does so with high reaction rates and potentially high efficiencies.^{3,4} An additional advantage of the solar redox cycle compared with other solar approaches is its ability to co-split H₂O and CO₂ simultaneously or separately and therefore control the quality (both purity and stoichiometry) of the syngas *in situ*, consequently obtaining a tailored mixture of H₂ and CO suitable for FT synthesis.⁵ This direct approach eliminates the energy penalty associated with additional refinement steps for adjusting the syngas mixture. In contrast, the electrolytic pathway

Context & scale

The aviation sector, which strongly relies on fossil-derived kerosene, is responsible for vast amounts of anthropogenic greenhouse gas emissions. To avoid these emissions, solar energy can be leveraged to efficiently produce sustainable drop-in fuels, e.g., solar-made synthetic kerosene, which is fully compatible with the existing global jet fuel infrastructures for its storage, distribution, and end-use in jet engines. This work advances the technological readiness level of solar fuels production by demonstrating the technical feasibility of the entire sun-to-liquid fuel process chain, from H₂O and CO₂ to kerosene, in a pilot-scale solar tower. We evaluate the performance of the solar reactor—the cornerstone technology—based on five primary metrics (namely, reaction selectivity, syngas quality, fuel purity, energy efficiency, and material stability) and experimentally validate its stable operation and full integration in the solar tower fuel plant.

(also called “power-to-X”)⁶ requires the production of substantial excess H₂ by water electrolysis using solar electricity that is subsequently consumed via the reverse water-gas shift reaction (RWGS reaction: H₂ + CO₂ = H₂O + CO, endothermic by 95.9 kJ/mol above 800°C) to obtain syngas suitable for FT synthesis. As will be shown in this study, the thermochemical approach bypasses the solar electricity generation, the electrolysis, and the RWGS steps, directly producing solar syngas of desired composition for FT synthesis, i.e., three steps are replaced by one.

Ceria (CeO₂) is currently considered the state-of-the-art redox material because of its rapid redox kinetics and long-term stability.⁷ The two-step thermochemical redox cycle is represented by:



where δ denotes the nonstoichiometry—a measure of the oxygen exchange capacity and therefore of the fuel yield per cycle. For typical operating conditions of the reduction step at 1,500°C and 0.1 mbar, and the oxidation step at 900°C and 1 bar, thermodynamics predict $\delta = 0.04$. Solar reactor concepts previously investigated for effecting the ceria redox cycle have included moving^{8–12} and stationary^{13–15} bulk structures, packed beds,^{16,17} moving beds,^{18,19} and aerosol flow^{20,21} of particles. Of special interest is the solar reactor concept based on a cavity-receiver containing reticulated porous ceramic (RPC) structures made of ceria,^{22,23} which provide efficient heat and mass transfer. Using an early prototype, the conversion of H₂O and CO₂ to renewable kerosene was demonstrated at the laboratory scale using a high-flux solar simulator.²⁴ Recently, two identical solar reactors were operated at the focus of a solar parabolic concentrator for performing both redox steps of the thermochemical cycle simultaneously by alternating the concentrated solar input between them.²⁵ While one solar reactor was performing the endothermic reduction step on sun, the second solar reactor was performing the exothermic oxidation step off sun, yielding a semi-continuous flow of syngas suitable for either methanol or FT synthesis. Stable outdoor operation was demonstrated for this solar fuel system, for which the mean solar radiative power input ($P_{\text{so-lar}}$) through the solar reactor’s aperture was 5 kW.²⁵

Despite recent advances, the scalability of the solar reactor remains a critical challenge to the commercialization of solar fuel production. The solar parabolic dish configuration is limited in size because of mechanical constraints due to wind and weight loads. Although multiple solar parabolic dishes may be deployed for scaling-up, a solar tower configuration features significant economy-of-scale advantages, as already seen for concentrated solar thermal power (CSP) plants,²⁶ and will likely be seen for solar fuel plants as well. Ultimately, the solar reactor technology will have to be scaled up for a solar tower configuration. Here, we describe the design, fabrication, and testing of a 50-kW solar reactor and experimentally demonstrate, for the first time, the entire sun-to-fuel process chain from H₂O and CO₂ to kerosene in a solar tower configuration. This pioneer demonstration was realized within the framework of the EU Horizon 2020 project SUN-to-LIQUID.²⁷ We evaluate and report the performance of the solar reactor—the cornerstone technology—based on five primary metrics, namely, reaction selectivity, syngas quality, fuel purity, energy efficiency, and material stability. The operation of a fully integrated solar tower fuel

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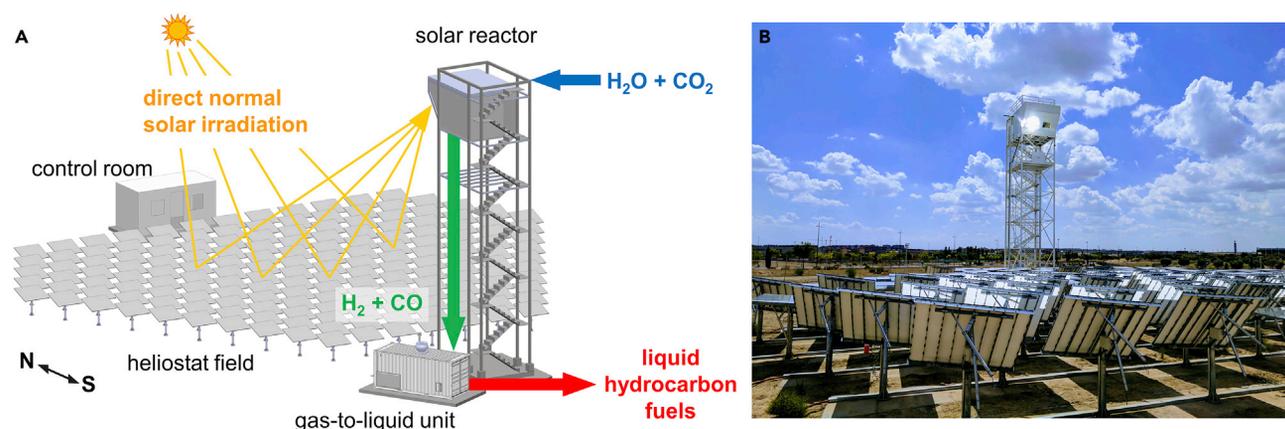


Figure 1. Overview of the solar tower fuel plant installed at IMDEA Energy (Spain)

(A) Schematic of the solar tower fuel plant, encompassing the solar tower concentrating facility, the solar reactor, and the GtL unit. A heliostat field concentrates the direct normal solar irradiation onto a solar reactor mounted on top of the solar tower. The solar reactor co-splits H_2O and CO_2 and produces a specific mixture of H_2 and CO (syngas), which in turn is processed to liquid hydrocarbon fuels using the FT-based GtL unit located next to the solar tower base. All sub-systems are operated from the control room.

(B) Photograph of the solar tower fuel plant during operation.

plant under intermittent solar radiation provides compelling evidence of the technical feasibility of the solar thermochemical technology for industrial scale implementation.

RESULTS AND DISCUSSION

The solar tower fuel plant, realized at IMDEA Energy in Spain, is depicted in Figure 1. It integrates three sub-systems: (1) the solar tower concentrating facility, (2) the solar reactor, and (3) the gas-to-liquid (GtL) unit. The solar concentrating facility consists of a solar tower with a south-facing heliostat field: an array of 169 sun-tracking spherical reflectors, each with an area of 3 m^2 , delivering a P_{solar} of about 50 kW into the 16-cm diameter aperture of the solar reactor, which corresponds to an average solar concentration ratio of approximately 2,500 suns, with a peak above 4,000 suns (1 sun is equivalent to a solar radiative flux of 1 kW/m^2).²⁸ The solar reactor is mounted on top of the solar tower at an optical height of 15 m, tilted 40° downward relative to the horizontal plane, and aimed at the power-weighted center of the heliostat field. On the ground next to the solar tower, the GtL unit is fully assembled inside a modular container. The experimental setup, peripheral components, and measurement instrumentation are described in detail in the supplemental information. The heliostat field is shown in the photograph of Figure S1. The solar reactor is described in experimental procedures.

An exemplary redox cycle operated in a temperature/pressure-swing mode is shown in Figure 2, where the nominal RPC temperature, the reactor pressure, and the gas product flow rates of O_2 , CO , and H_2 are plotted as a function of time. The experimental conditions and results of this run are summarized in Table 1. During the reduction step at an average $P_{\text{solar}} = 42.0 \pm 6.2 \text{ kW}$, and under vacuum conditions, the nominal RPC temperature rapidly increased up to the reduction end temperature ($T_{\text{reduction, end}}$) of $1,502^\circ\text{C}$ at a mean heating rate of about $100^\circ\text{C min}^{-1}$. Accordingly, the rate of O_2 evolution increased to a maximum of $8.7 \pm 0.2 \text{ L min}^{-1}$. Integrated over the entire reduction step, a total amount of $36.2 \pm 0.7 \text{ L O}_2$ was released, which, assuming all ceria reacted uniformly, corresponds to a specific oxygen exchange capacity of 0.002 L/g ceria and an average oxygen nonstoichiometry

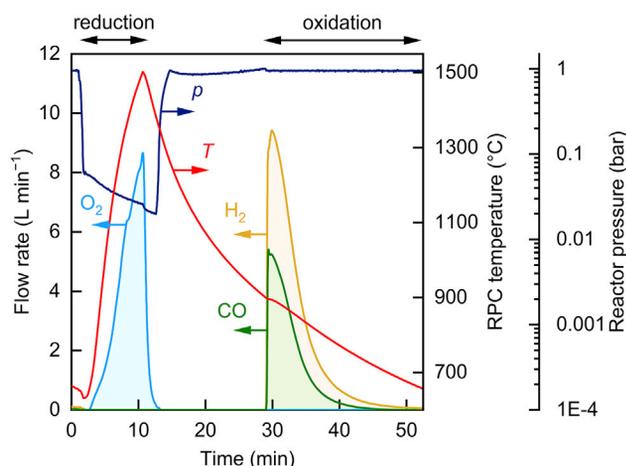


Figure 2. Temporal variations of the nominal RPC temperature, reactor pressure, and gaseous product (O₂, CO, and H₂) evolution rates during an exemplary redox cycle

Experimental conditions during reduction: mean $P_{\text{solar}} = 42.0 \pm 6.2$ kW; volumetric flow rate of Argon (V_{Ar}) = 5.0 L min⁻¹ at pressure (p) ≤ 70 mbar. Experimental conditions during oxidation: $\dot{n}_{\text{H}_2\text{O}} = 0.033$ mol s⁻¹, $\dot{n}_{\text{CO}_2} = 0.0074$ mol s⁻¹, at $p \approx 1$ bar. Ceria RPC mass (m_{RPC}) = 18.1 kg.

at the end of the reduction step of $\delta = 0.031$. This indicates that the system approached thermodynamic equilibrium, consistent with previous tests with the laboratory-scale reactor.²² At the end of the reduction step after 8.8 min, the solar input was interrupted ($P_{\text{solar}} = 0$), and the oxygen release rate rapidly decreased to zero, whereas the RPC naturally cooled down to the nominal oxidation start temperature ($T_{\text{oxidation,start}}$) of 900°C within 18.3 min. Oxidation was initiated by simultaneously injecting H₂O and CO₂ at molar flow rates of $\dot{n}_{\text{H}_2\text{O}} = 0.033$ mol s⁻¹ and $\dot{n}_{\text{CO}_2} = 0.0074$ mol s⁻¹. Both H₂ and CO production rates peaked shortly after at 9.4 ± 0.8 L min⁻¹ and 5.4 ± 0.4 L min⁻¹, respectively, and decreased monotonically until the ceria was fully re-oxidized after 24.0 min when the oxidation end temperature ($T_{\text{oxidation,end}}$) reached 654°C. Integrated over the entire oxidation period, a total amount of 48.9 ± 3.9 L H₂ and 24.4 ± 2.0 L CO was produced. Mass balance of both redox steps yields a corresponding molar ratio (H₂ + CO):O₂ = 2.03 ± 0.21 , indicating full selectivity for the conversion of H₂O to H₂ and CO₂ to CO. No side reactions or by-products were detected. Note that the molar ratio of the fed reactants reached H₂O:CO₂ = 4.5 because excess water was required to obtain the desired syngas quality for FT synthesis. For the exemplary run of Figure 2, H₂:CO = 2.01 ± 0.35 , which is suitable for FT synthesis.

Besides reaction selectivity and syngas quality, an important performance indicator that particularly affects the economic viability of the process is the solar-to-syngas energy conversion efficiency ($\eta_{\text{solar-to-syngas}}$), defined as the ratio of the calorific value of the syngas produced over the cycle to the sum of solar radiative energy input (Q_{solar} , obtained by integrating P_{solar} over the cycle, $Q_{\text{solar}} = \int P_{\text{solar}} dt$) and additional parasitic energy inputs associated with inert gas consumption and vacuum pumping (see supplemental information for efficiency formulation; Figures S2 and S3 for details on the solar radiative power determination). The energy conversion efficiency depends primarily on the amount of syngas produced (H₂ and/or CO) during the oxidation step, compared with the amount of solar energy required to release O₂ during the reduction step. For the exemplary run of Figure 2, $\eta_{\text{solar-to-syngas}} = 4.1 \pm 0.8\%$ at an average $P_{\text{solar}} = 42.0 \pm 6.2$ kW. For pure CO₂-splitting, $\eta_{\text{solar-to-syngas}} = 5.6 \pm 1.0\%$ at an average $P_{\text{solar}} = 55.8 \pm 8.2$ kW. From an operational perspective,

Table 1. Experimental conditions and results of the exemplary solar redox cycle of Figure 2

Variable	Symbol	Value	Unit
Ceria RPC mass	m_{RPC}	18.1	kg
Average solar power input during reduction	P_{solar}	42.0 ± 6.2	kW
Solar power input during oxidation	N/A	0	kW
Reduction start temperature	$T_{\text{reduction,start}}$	632	°C
Reduction end temperature	$T_{\text{reduction,end}}$	1,502	°C
Oxidation start temperature	$T_{\text{oxidation,start}}$	900	°C
Oxidation end temperature	$T_{\text{oxidation,end}}$	654	°C
Ar flow rate during reduction	V_{Ar}	5.0	L min ⁻¹
H ₂ O flow rate during oxidation	$\dot{n}_{\text{H}_2\text{O}}$	0.033	mol s ⁻¹
CO ₂ flow rate during oxidation	\dot{n}_{CO_2}	0.0074	mol s ⁻¹
Reactor pressure during reduction	N/A	26–70	mbar
Reactor pressure during oxidation	N/A	atmospheric	N/A
Reduction duration	N/A	8.8	min
Duration of cooling-down	N/A	18.3	min
Oxidation duration	N/A	24.0	min
Cycle duration	N/A	51.1	min
Mean heating rate	N/A	98.9	°C min ⁻¹
Peak O ₂ evolution rate	N/A	8.7 ± 0.2	L min ⁻¹
Total amount of O ₂ released	N/A	36.2 ± 0.7	L
Average nonstoichiometry of ceria after reduction	δ	0.031 ± 0.001	N/A
Peak H ₂ O evolution rate	N/A	9.4 ± 0.8	L min ⁻¹
Total amount of H ₂ O produced	N/A	48.9 ± 3.9	L
Peak CO evolution rate	N/A	5.4 ± 0.4	L min ⁻¹
Total amount of CO produced	N/A	24.4 ± 2.0	L
Molar ratio (H ₂ + CO)/O ₂	N/A	2.03 ± 0.21	N/A
Molar ratio H ₂ /CO	N/A	2.01 ± 0.35	N/A
Solar-to-syngas energy efficiency	$\eta_{\text{solar-to-syngas}}$	4.1 ± 0.8	%

the primary difference between these two reported efficiencies was P_{solar} . A higher P_{solar} for the pure CO₂-splitting run resulted in rapid heating and a shorter reduction cycle, which in turn led to lower Q_{solar} ($Q_{\text{solar}} = \int P_{\text{solar}} dt = 20.1$ MJ, versus 22.2 MJ for the co-splitting of H₂O and CO₂) and consequently higher $\eta_{\text{solar-to-syngas}}$. On the other hand, the co-splitting run used excess water, which consumed part of Q_{solar} upon heating to $T_{\text{reduction,end}}$ and led to lower $\eta_{\text{solar-to-syngas}}$. Splitting pure H₂O and pure CO₂ in separate cycles and mixing the product gases H₂ and CO can also be applied to obtain the syngas composition required for FT synthesis, eliminating the need for excess water during a co-splitting run.

These measured values of energy conversion efficiency were obtained without any implementation of heat recovery. Specifically, the sensible heat rejected during the temperature-swing redox cycling accounted for more than 50% of Q_{solar} . This fraction can be partially recovered via thermochemical heat storage, as demonstrated with a packed bed of Al₂O₃ spheres, which was able to recover half of the sensible energy stored for a temperature swing between 1,400°C and 900°C.²⁹ Thermodynamic analyses indicate that sensible heat recovery could potentially boost $\eta_{\text{solar-to-syngas}}$ to values exceeding 20%.^{3,4} Furthermore, it was evident from the temperature distribution across the RPC that the reaction extent was not uniform. Heat-transfer modeling estimated a temperature difference between the directly irradiated front and the back surface of the ceria RPC to exceed 200°C.³⁰ This is mainly caused by the exponential decay of transmitted radiation (Bouguer's law) observed for a RPC of uniform porosity, resulting in a significant temperature gradient across the RPC thickness. The ratio between the actual released O₂ and the amount of O₂

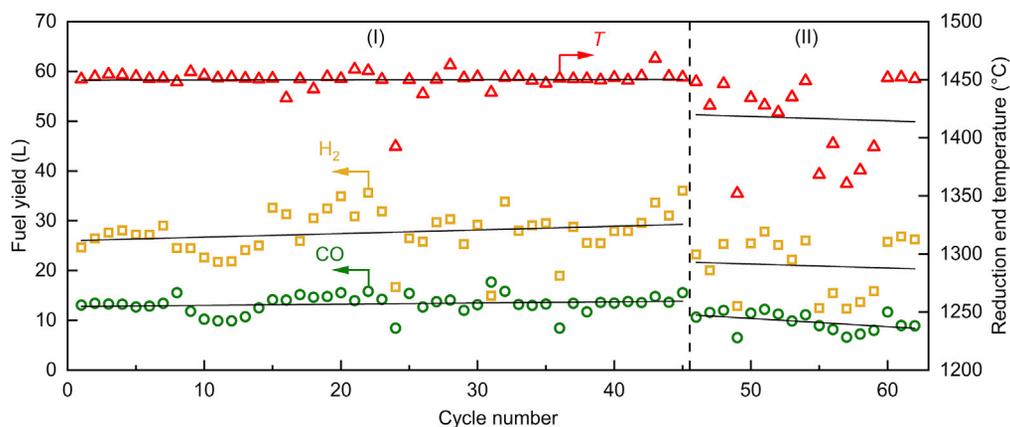


Figure 3. Multiple consecutive redox cycles

Nominal ceria RPC temperature at the end of the reduction step and total amounts of produced H_2 and CO per cycle for 62 consecutive redox cycles, yielding $5,191 \pm 364$ L of syngas with a composition $31.8\% \pm 3.2\%$ H_2 , $15.2\% \pm 2.4\%$ CO , and $53.0\% \pm 3.6\%$ CO_2 (H_2O condensed). Linear fits are shown. L denotes standard liters.

that could theoretically be released if all ceria mass would have reached uniform temperature at the end of the reduction step is estimated to be approximately 0.5. This ratio can be increased by modifying the radiation attenuation, for example, by manufacturing hierarchically ordered porous structures with a step-wise porosity gradient, which can augment the volumetric radiative absorption and lead to a more uniform temperature distribution and, ultimately, higher efficiencies.^{31,32}

Note that $\eta_{\text{solar-to-syngas}}$ only considers the performance of the solar reactor sub-system. The energy efficiency of the entire solar fuel plant should also consider the performance of the other two sub-systems upstream and downstream of the solar reactor, namely, the optical efficiency of the solar concentrating tower facility (η_{optical}) and the energy efficiency of the GtL unit (η_{GtL}). η_{optical} depends on the heliostat layout, geometry, reflectivity, tracking accuracy, shading/blocking, attenuation, and cosine losses and can reach values up to 70% while keeping a mean solar flux concentration of 2,500 suns over the solar reactor's aperture, provided radiation spillage is collected and used (for example to preheat gaseous reactants).²⁸ η_{GtL} depends mainly on the targeted product, catalyst, and syngas composition. When targeting methanol synthesis and assuming autothermal operation, 90% mass conversion, and accounting for the equivalent thermal energy penalty for syngas compression to 60 bars, η_{GtL} was estimated to be 75%.²⁵ When targeting FT synthesis, η_{GtL} further depends on the definition of mass conversion, since several valuable products (e.g., kerosene or diesel) can be co-generated.

Stable performance of the solar reactor over a large number of redox cycles is essential for any potential commercial application. The morphological stability of a similar ceria RPC was previously demonstrated with 227 consecutive redox cycles in a 4-kW solar reactor²⁴ and with 500 consecutive cycles in an infrared.²² For the 50-kW solar reactor in this study, 62 consecutive redox cycles were performed during a dedicated and continuous fuel production campaign. A representative cycle is shown in Figure S4. The cycles were conducted over a period of 9 days, 6–8 cycles/day (except for one day when cycle #24 was interrupted by clouds), with an average duration of 53 min/cycle and a total experimental time of 55 h (see also the operational strategy described in Figure S5 during a representative day run, including a heating phase, a pre-cycle, consecutive cycling, and a natural cooling phase). Figure 3 shows

the nominal RPC temperature at the end of the reduction step and the total amounts of H₂ and CO produced per cycle for all 62 cycles. During the first 45 cycles (region I), the targeted $T_{\text{reduction, end}}$ of $1,450^{\circ}\text{C} \pm 18^{\circ}\text{C}$ was reached for all cycles (except for cycle #24), yielding a relatively constant fuel production. However, during the last 17 cycles (region II), $T_{\text{reduction, end}}$ varied as several cycles were stopped early due to critical high temperatures ($>1,500^{\circ}\text{C}$) measured at the back of the RPC cavity. These temperature variations from cycle to cycle directly resulted in variations of the oxygen released and, consequently, the fuel amounts produced. Although an effort was made to maintain constant operating conditions for all consecutive cycles, temporal variations of the direct normal irradiance (DNI) and of the tracking of the heliostat field resulted in varying P_{solar} and, consequently, in temperature and product gas fluctuations. In more than 90% of the cycles, the trend in CO and H₂ yield was as expected, i.e., increasing together or decreasing together with higher or lower reduction temperatures, respectively. For the few cycles where the expected trend is not observed, the deviation is minimal, presumably caused by temporal and/or spatial variations of the RPC temperature affecting the reduction extent of ceria (δ) and in turn its oxidation with H₂O and CO₂. Degradation of the ceria RPC caused by the local formation of cracks was observed (see [supplemental information](#), in particular [Figures S6](#) and [S7](#)), presumably caused by the critical temperatures measured at the back of the RPC cavity. Nonetheless, the interlocking design of the RPC bricks ensured the integrity of the cavity assembly. Overall, $5,191 \pm 364$ L of syngas were produced with a composition of $31.8\% \pm 3.2\%$ H₂, $15.2\% \pm 2.4\%$ CO, and $53.0\% \pm 3.6\%$ unreacted CO₂, whereas the unreacted H₂O was condensed. The corresponding molar ratio of H₂:CO was 2.1. Around 91% of the produced syngas was subsequently processed on-site by the GtL unit, yielding a liquid phase containing 16% kerosene and 40% diesel, and a wax phase containing 7% kerosene and 40% diesel. See [Figure S8](#) for additional details on the FT product distribution.

In summary, the technical feasibility of the entire thermochemical process chain to produce solar liquid hydrocarbon fuels from H₂O and CO₂ has been demonstrated with a pilot-scale solar tower fuel plant that integrates, in series, the three main sub-systems, namely: the solar concentrating tower, the solar reactor, and the GtL unit. The solar reactor produced syngas with selectivity, purity, and quality suitable for FT synthesis. Although the $\eta_{\text{solar-to-syngas}}$ is still in the single digits, it has the potential to reach competitive values of over 20% by recovering rejected heat during the temperature-swing redox cycle and by improving the volumetric absorption of the porous structures. The ceria RPC remains the most critical component of the solar reactor and further progress with the manufacturing of mechanically robust porous structures remains essential. Alternative material compositions, e.g., perovskites³³ or aluminates,³⁴ may yield sufficient redox capacity at lower, more moderate temperatures or under isothermal conditions. Adjustments to the cavity geometry and concentrating optical system, i.e., by incorporating a secondary compound parabolic concentrator (CPC), can further improve the uniformity of the radiative flux distribution within the cavity and consequently alleviate the thermal stressing. One approach to scaling up the solar fuel plant would be to use an array of solar cavity-receiver modules arranged side-by-side, each attached to hexagon-shaped CPC in a honeycomb configuration. The solar tower fuel plant described here represents a viable pathway to global-scale implementation of solar fuel production. If CO₂ is further captured from the air or derived from a biogenic source, the resulting drop-in hydrocarbon fuels, e.g., kerosene, can be considered carbon neutral.^{25,35} Life-cycle assessment and economic feasibility of the complete fuel process chain, analogous to the pathway demonstrated in this study, as well as benchmarking

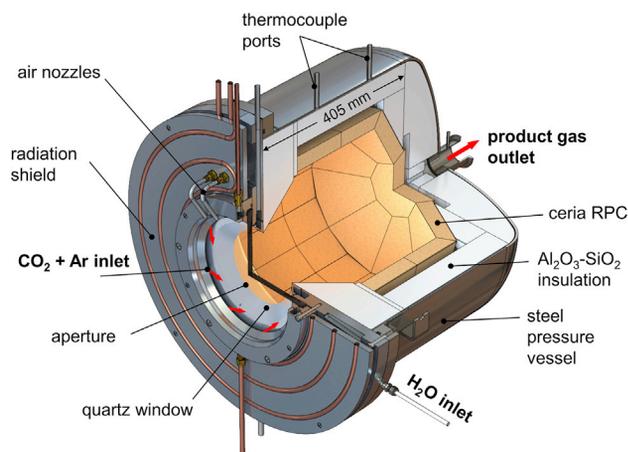


Figure 4. Schematic of the solar reactor for splitting H_2O and CO_2 via the ceria-based thermochemical redox cycle

It consists of a cavity-receiver containing a ceria RPC structure directly exposed to concentrated solar radiation entering through a windowed circular aperture. During the reduction step, the RPC is exposed to the high solar fluxes; O_2 evolves. During the oxidation step, reacting gases CO_2 and H_2O enter via tangential inlet ports at the front and flow across the porous RPC; syngas is formed. Product gases (O_2 during the reduction step, syngas during the oxidation step) exit via an axial port at the rear of the vessel.

vis-à-vis alternative approaches to the production of drop-in fuels using solar energy, were discussed in previous publications.^{25,36,37}

EXPERIMENTAL PROCEDURES

Resource availability

Lead contact

Further information and requests for resources should be directed to Aldo Steinfeld, aldo.steinfeld@ethz.ch.

Materials availability

This study did not generate new unique materials.

Data and code availability

The main data supporting the findings of this study are available within the paper and its supporting documentation. Source data are available with this paper.

The solar reactor was based on a previous laboratory-scale design,²² which was scaled up from 4 kW to a nominal 50 kW of P_{solar} , which corresponds to a scaling factor of 12.5. Its configuration is schematically shown in Figure 4. It consists of a well-insulated cavity-receiver with a 16-cm diameter circular aperture where concentrated solar radiation enters. The aperture is sealed with a transparent quartz window mounted on a refrigerated radiation shield and actively cooled from the outside by air nozzles. The cavity contains a cylindrical structure of interlocking RPC bricks made of ceria (see Figure S9). With this arrangement, the RPC bricks are directly exposed to concentrated solar radiation coming from the heliostat field, providing efficient radiative heat transfer directly to the reaction site. During the oxidation step, reacting gases CO_2 (purity 99.9%) and H_2O (deionized) enter the reactor via tangential inlet ports at the front and flow across the porous RPC; product gases (O_2 during the reduction step, syngas during the oxidation step) exit via an axial port at the rear of the vessel. A lower purity of CO_2 feedstock, i.e., containing 1%–2% air as

might be obtained by direct air capture, would not significantly influence the performance of the solar reactor because N_2 is inert and O_2 would be consumed by oxidizing the reduced ceria RPC.²⁵ A detailed process flow schematic is shown in Figure S10.

The solar reactor geometry was determined by applying CFD simulations.^{30,38} Key scaling parameters and considerations when moving from the 4-kW lab-scale prototype²² to the 50-kW reactor design included: (1) determining the aperture size paired to a given heliostat field in order to achieve a mean solar flux over the aperture of 2,500 suns; (2) selecting a cavity geometry that gives an apparent absorptivity approaching 1; (3) determining the RPC exposed surface area to maintain an incident flux of 125 suns; (4) arranging the inlet/outlet gas ports to achieve uniform and stable fluid flow across the RPC; (5) increasing the RPC thickness and number of facets to support a larger interlocking brick structure; and (6) maintaining the RPC porosity without sacrificing structural integrity. The dual-scale interconnected porosity (mm and μm size pores) provided volumetric radiative absorption during the reduction step and faster reaction kinetics during the oxidation step.³⁹ Engineering details are provided in the [supplemental information](#).

Solar-produced syngas exits the solar reactor sub-system at the top of the tower, and after condensing unreacted H_2O and passing through in-line gas analysis, flows at near ambient pressure down the tower, where it is pressurized and stored in a 50-L buffer tank at 30–150 bar. The GtL unit controller automatically draws syngas from the buffer tank to perform the FT catalytic conversion in its cobalt-based packed-bed reactor at 30 bar and 210°C. The FT synthesis requires a syngas with $H_2:CO$ molar ratio of around 2.15,⁴⁰ which the solar reactor sub-system is able to match very closely by varying the mass flow rate of reactants H_2O and CO_2 during the oxidation step. The resulting long-chain hydrocarbons are collected in a downstream vessel for sampling and analysis. Despite the intermittent nature of the solar resource, the buffer tank enables the GtL unit to be operated with any desired production schedule, ranging from 24/7 slow and steady operation to short duration and high production rate operation.

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at <https://doi.org/10.1016/j.joule.2022.06.012>.

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AUTHOR CONTRIBUTIONS

S.Z., E.K., P.H., and A. Steinfeld contributed to the solar reactor design; S.Z., E.K., D.N., M.S., and A.P. assembled the experimental setup and executed the experiments; M.R. and J.G.-A. managed the realization of the solar concentrating tower

facility; D.L. and E.d.W. operated the GtL unit; S.B. contributed to the implementation of the solar flux measurement instrumentation; A. Sizmann coordinated the EU project SUN-to-LIQUID; and A. Steinfeld wrote the manuscript with input from all authors.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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- **Bauhaus Luftfahrt e.V. (Germany)**
- **DLR-German Aerospace Center (Germany)**
- **HyGear Technology and Services B.V. (the Netherlands)**
- **Abengoa Research SL (Spain)**
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Aldo Steinfeld (PhD University of Minnesota, 1989) is Full Professor of the Department of Mechanical and Process Engineering at ETH Zurich, where he holds the Chair of Renewable Energy Carriers. From 2004-2014 he directed the Solar Technology Laboratory at the Paul Scherrer Institute. At ETH, he served as the Head of the Institute of Energy Technology (2005-2007) and Associate Head of Research of the Department of Mechanical and Process Engineering (2007-2009). He was member of the Board of Directors of the International Solar Energy Society (2015-2019). For SOLARPACES, he served as the Operating Agent of Task-II Solar Chemistry (1996-2004) and organized and chaired the SOLAPACES Conference 2002 in Zurich.

His research program is aimed at the advancement of the thermal and chemical engineering sciences applied to renewable energy technologies. His fundamental research focusses on high-temperature heat/mass transfer phenomena, multi-phase reacting flows, thermochemistry and functional redox materials. These are applied in the development of technologies for concentrated solar power and solar fuels production, solar-driven thermochemical processing of energy-intensive chemical commodities, direct air capture of CO₂ and its utilization, energy storage and sustainable energy systems.

Prof. Steinfeld was the Editor-in-Chief of the ASME Journal of Solar Energy Engineering (2005-2009), co-Editor of the CRC Handbook of Hydrogen Energy (2014), and is currently serving in several editorial boards. He has authored over 350 refereed journal articles (citation h-index = 94) and filed 25 patents. His contributions to science and education have been recognized with the ASME Rice Award (2006), the Yellott Award (2008), the European Research Council Advanced Grant (2012), the ISES Farrington Daniels Award (2013), the Heat Transfer Memorial Award (2013), the ASME Kreith Energy Award (2016); and the SOLARIS Life-Long Contribution Award by Japan's Heat Transfer Society (2021). Two spin-offs emerged from his research group: Climeworks commercializes the technology for CO₂ capture from air, and Synhelion commercializes the technology for solar fuel production. Prof. Steinfeld is member of the Swiss Academy of Engineering Sciences and the Pan-American Academy of Engineering.

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Dr. Andreas Sizmann is Head of "Future Technologies and Ecology of Aviation". He joined Bauhaus Luftfahrt, a national think tank for the long-term future of mobility and aviation, in 2007. He established future innovation potential assessment based on latest advancements in science and technology, successively in the methodology of future technology analysis, and in the fields of energy technologies, alternative fuels, materials, information and communication technologies. Since 2011 he also serves as member of the Working Group "Protecting the Environment and the Energy Supply" of the "Advisory Council for Aviation Research and Innovation in Europe (ACARE)" and was member of the Expert Group of "Sustainable Alternative Fuels for Aviation (SUSTAF)" of the "International Civil Aviation Organization (ICAO)" in 2014. Prior to his role at Bauhaus Luftfahrt Dr. Sizmann was, among other assignments, Director of Optical Research at Innovance Networks (Ottawa, Canada), professor on temporary assignment at Ludwig-Maximilians University Munich and University of Constance, guest scientist at the IBM Almaden Research Center (San Jose, CA, USA) and at the Laboratoire Kastler Brossel de l'École Normale Supérieure (Paris, France) as well as research scientist at the MPI for Quantum Optics in Garching. Dr. Sizmann received several degrees in physics, the Habilitation at the Friedrich-Alexander University Erlangen (1999), the Doctorate degree of the Ludwig-Maximilians University Munich (1992) and the Diploma in physics "with distinction" of the Technical University of Munich (1989). He is member of the German Physical Society (DPG).