Modelling of CO_2 conversion by atmospheric pressure microwave plasma enhanced by concentrated solar radiation

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Abstract: The combined use of plasma and concentrated solar radiation can lead to more viable CO_2 conversion. The modelling of an experimentally-evaluated solar-microwave plasma reactor is presented. The model comprises fluid flow, heat transfer, chemical kinetics, electrons and heavy-species energy conservation, electrostatics/microwave transport, and radiative transport. Modelling results indicate that increased conversion is caused by the greater power density of the plasma due to the absorption of solar radiation.

Keywords: CO_2 decomposition, solar fuels, chemical synthesis, sustainable chemicals

1. Introduction

The production of chemicals and fuels using renewable energy via environmentally-benign methods is essential for achieving sustainable development. Particularly, the valorization of carbon dioxide (CO_2), the main driver of climate change, utilizing solar irradiation or electricity from renewable energy sources, can lead to economic development and mitigation of CO_2 emissions. The novel concept of solar-plasma chemical conversion aims at the integrated utilization of solar power and renewable electricity to attain the sustainability advantages and scalability of concentrated solar thermochemical techniques together with the versatility, continuous operation, and high-efficiency of plasmachemical methods. A review of CO_2 conversion by solar-plasma reactors, including reactor design approaches and actual implementations, is presented in [1].

Solar-plasma reactors, as schematically depicted in **Fig 1a**, utilize an influx of concentrated solar radiation together with electric power to sustain plasma and drive chemical conversion kinetics. Solar-plasma processes are characterized by the ratio of nominal input solar power (P_s) to nominal input electric power (P_e) as either Plasma-Enhanced Solar thermochemical (PES, for $P_s > P_e$) or Solar-Enhanced Plasmachemical (SEP, for $P_e > P_s$). (Actual solar-plasma reactor operation would experience fluctuating P_s and/or P_e.) A representative PES approach is CO_2 conversion by the solar-gliding arc reactor devised by Nagassou *et al.* for P_s up to 525 W and P_e up to 240 W [2]. The relatively simple design and operation of the gliding arc makes it particularly suitable for its integration in solar thermochemical processes. An implementation of a SEP approach is the Solar-Enhanced Microwave Plasma (SEMP) CO_2 conversion by Mohsenian *et al.* [3, 4]. The SEMP reactor is designed for P_s up to 525 W and P_e up to 1250 W, and its computational modeling is the focus of the present work.

2. Solar-Enhanced Microwave Plasma (SEMP)

SEMP approaches can lead to greater conversion and energy efficiency thanks to increased power density of the plasma due to the absorption of solar radiation. This is possible given that, although CO_2 in thermal equilibrium is largely transparent to solar radiation, CO_2 in thermal nonequilibrium (i.e., dissimilar electron and heavy-species temperatures, characteristic of nonthermal plasma) can present significant CO_2 absorption. The absorption of solar radiation by CO_2 in nonthermal plasma state has been first studied using radiative properties calculated by the line-by-line radiative transport code SPARK by Lino Da Silva and collaborators [5] within a one-dimensional reactor model in [6]. The study shows that solar radiation absorption drastically increases with increasing degree of thermal nonequilibrium, but so does radiative emission. The net amount of radiative power absorbed by plasma is therefore tightly dependent on the characteristics of the reactor design and the operating conditions of the process.

The design of the SEMP reactor experimentally-characterized in [3, 4] is schematically shown in **Fig. 1b**. The design of solar-plasma reactors is intrinsically dependent on the source of solar radiation (i.e., on the intensity of incident solar irradiation, as well as on the distribution and directionality of radiative intensity). The SEMP reactor is designed to receive radiation from a 6.5 kW high-flux solar simulator fitted with a parabolic concentrator. The focal point of the incident radiation is located at the center of the intersection between the discharge tube and a tapered waveguide. The measured maximum solar power into the reactor is P_s = 525 W. The reactor is powered by a P_e = 1.25 kW magnetron operating at 2.45 GHz. The working gas is injected tangentially to produce swirl flow that helps stabilize the plasma. The discharge tube is made of boron nitride and is surrounded by a cut-off tube to mitigate microwave losses.

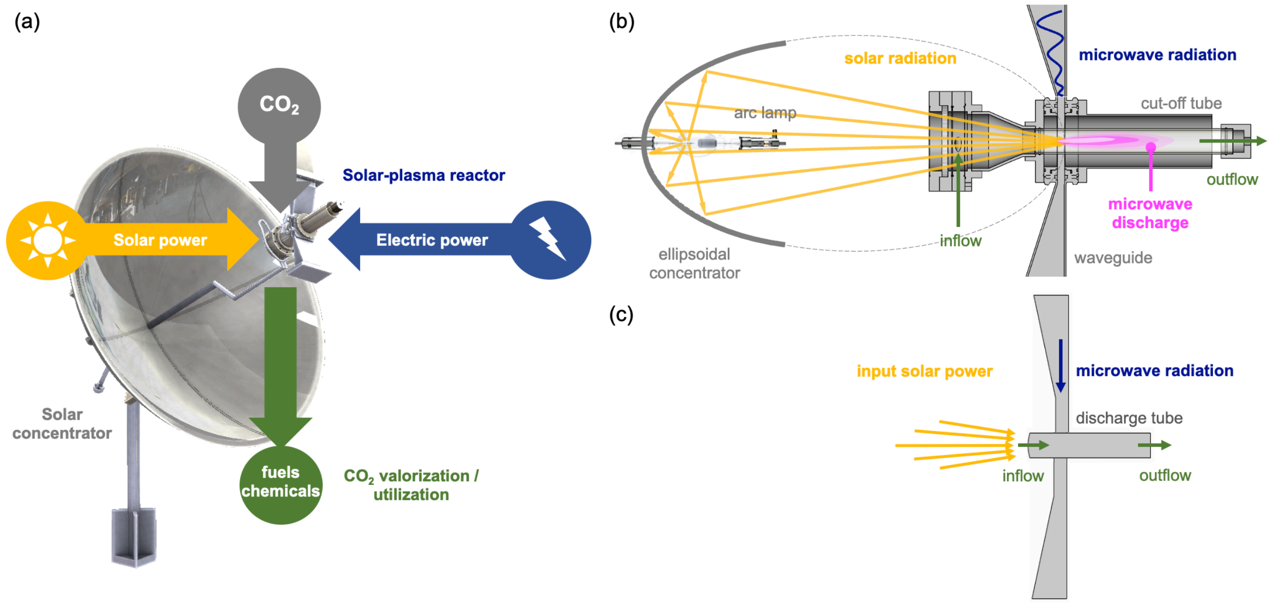

Fig. 1. Solar-plasma chemical conversion. (a) Solar-plasma chemical conversion seeks to exploit the advantages of solar thermochemical and plasmachemical approaches towards the sustainable synthesis of fuels and chemicals. (b) Design schematic of an experimentally-evaluated solar-enhanced microwave plasma (SEMP) reactor depicting the incidence of radiation from a high-flux solar simulator and its interaction with microwave plasma. (c) Spatial domain of the computational SEMP reactor model encompassing the waveguide and the discharge tube.

3. Modelling of SEMP CO_2 conversion

A primary aspect of the computational modelling of the SEMP reactor is the definition of the computational domain. To appropriately analyse microwave discharges, the computational domain has to encompass not only the plasma region (e.g., discharge tube), but also the region of propagation of microwaves (e.g., waveguide). The two-dimensional computational domain for the analysis of the SEMP reactor is presented in **Fig. 1c**. The waveguide regions allow the description of the incident and transmitted microwave power. The discharge tube domain includes a curved open region upstream over which the inflow gas is imposed. The curvature of the inflow boundary follows the parabolic profile from the incident solar radiation. Therefore, the incident solar radiation can be imposed as being normal to the inflow boundary and with magnitude such that the total incident radiation integrated over that boundary is equal to the input solar power P_s.

The SEMP reactor model utilizes a fully-coupled approach to solve the equations comprising the description of fluid flow, heat transfer, Ar-CO_2 chemical kinetics, energy conservation for electrons and heavy-species, electrostatics, and radiative transport in participating media through the discharge tube, together with the description of the microwave electromagnetic field through the waveguide. The Ar-CO_2 chemical kinetics model includes 12 species, 3 argon-related species (Ar, Ar^*, Ar^+) and 9 CO_2-related species (CO_2, CO, CO_2^+, O_2, O_2^-, O_3, O^-, O, and C), which interact through a total of 59 reactions.

An important aspect of the model is the description of the interaction of solar radiation with plasma. Such description requires the modeling of radiative transport in participating media. This is accomplished by solving the Radiative Transport Equation (RTE). Due to the high computational cost of the simulations, the plasma is considered a gray medium (i.e., wavelength-independent radiative properties) with a gray absorption coefficient function of both, electron and heavy-species temperatures, T_e and T_h, respectively. The absorption coefficient is calculated by integrating over wavelengths from 380 nm (ultra-violet) to 700 nm (infra-red) the spectral absorption coefficient computed using SPARK [5] considering 34 types of atomic and molecular transitions ($O(10^6)$ transitions total). To appropriately resolve the directional dependency of radiation while limiting the total cost of the simulations, 12 directions are utilized.

Representative results of the simulation of the SEMP reactor, with and without input solar power, are presented in **Fig. 2**. It is to be noted that, even in the absence of incident solar power, the solution of the RTE produces a more accurate description of heat transfer within the discharge tube. This can be appreciated by the distribution of incident radiation I_G, defined as the integral of radiative intensity over all directions. The results show that the incorporation of solar power leads to greater temperatures and to greater CO_2 conversion. The results corroborate the expectation that the absorption of solar radiation allows the increase in power density by the plasma circumventing skin-depth limitations generally associated with the absorption of microwave power.

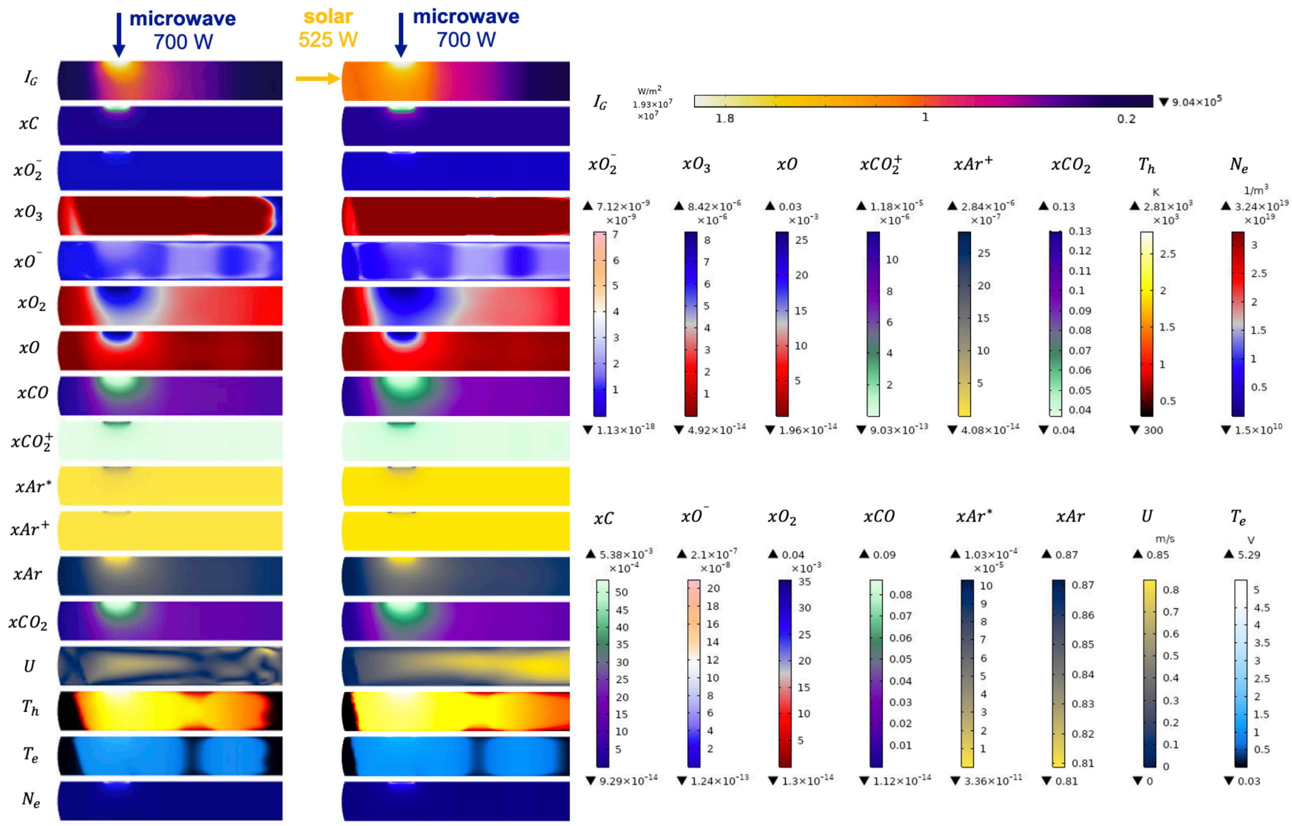

Fig. 2. Simulation results of SEMP reactor operation. Distribution of properties across the SEMP reactor chamber under microwave power-only operation (700 W incident microwave power) and under solar-enhanced operation (700 W of microwave power and 525 W of solar power), at 1 atm and 8 slpm of Ar-CO_2 (7:1 by volume). Incident radiation I_G, mole fraction x of the species included in the model (C, O_2^-, O_3, O^-, O_2, O, CO, CO_2^+, Ar^*, Ar^+, Ar, and CO_2), velocity U, heavy-species temperature T_h, electron temperature T_e, and electron number density N_e.

4. Conclusion

Solar-Enhanced Microwave Plasma (SEMP), by combining the advantages of solar thermochemical and plasmachemical processes, aims to lead to more viable CO_2 conversion. An experimentally-evaluated SEMP reactor is modeled using a fully-coupled 2D model encompassing the description of fluid flow, heat transfer, energy conservation for electrons and heavy-species, radiative transport in participating media, and Ar-CO_2 chemical kinetics through the discharge tube and waveguide. Increased conversion is caused by the greater power density due to the absorption of solar radiation.

Acknowledgements

The authors acknowledge financial support from the U.S. National Science Foundation award CBET-1552037.

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