Enhanced Carbon Dioxide Conversion by Atmospheric Pressure Microwave Plasma - Solar Processing

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Abstract: The conversion of CO_2 using renewable energy can help mitigate environmental emission and address global fuel needs. Solar thermochemical processes have the greatest sustainability potential, but are limited by the low reactivity of CO_2 and the intermittency of solar radiation. In contrast, plasma processes have the potential for high efficiency and continuous operation. The synergistic combination of microwave plasma and concentrated solar is investigated as a novel approach for greater viability CO_2 conversion.

Keywords: microwave plasma, solar thermochemistry, CO_2 decomposition.

1. Introduction

In the last decades, human activities have increased the concentration of carbon dioxide (CO_2) in the earth's atmosphere. Among the several methods to control the rising atmospheric CO_2 concentration, the decomposition of CO_2 to carbon monoxide (CO), a base chemical, not only can play a key role in curving greenhouse emissions but also in addressing the global reliance on liquid fuels. In recent years, microwave plasma CO_2 decomposition processes have been gaining greater attention due to their reliance on electricity, potentially from renewable sources, suitability for gas-phase processing, and potential for high efficiency. The latter is due to their potential for step-wise vibrational excitation, an effective channel for dissociation, as well as their relatively high heavy-species temperature, which promotes thermal dissociation [1-8].

Solar thermochemical processes directly utilize solar energy, the most abundant form of renewable energy, and hence present the greatest sustainability potential. However, the effectiveness of solar thermochemical processes is limited by the low reactivity of most feedstock gases to solar radiation, and by the inherent intermittency of the solar resource. Solar thermochemical synthesis processes rely on a catalytic medium to absorb solar radiation and drive heterogeneous thermochemical reactions [9-11]. The complexity of advanced catalytic systems together with the high intermittency of solar radiation often lead to high capital and operating costs.

Microwave Plasma Enhanced Solar (MPES) is investigates as a novel approach that seeks to combine the advantages of solar thermochemical and plasmachemical processes to enhance the efficiency and resiliency, and consequently improve the viability, of CO_2 decomposition processes. MPES processes use solar radiation to enhance the energy deposited in a microwave plasma to promote molecular decomposition kinetics. Moreover, MPES processes can use electrical energy to compensate for solar fluctuations or even operate solely using electrical energy, e.g. during night time when solar energy is not available, and are therefore suitable for continuous operation, a characteristic that can lead to significantly lower operational and capital costs.

2. Reactor Design and Rationale

In the MPES reactor, an influx of concentrated solar radiation is aimed to interact with a stable microwave plasma. Figure 1 schematically depicts the conceptual framework of a solar-plasma reactor system consisting of a solar concentrator to direct the solar radiation into the reactor, a microwave plasma applicator, and gas delivery system. The plasma applicator consists of a tapered rectangular wave guide, a dielectric discharge tube surrounded by a conducting cut-off tube, and a conical chamber to transfer the incoming solar radiation to the plasma generation point. This reactor is aimed to convert feedstock gases such as CO_2, CH_4, H_2O, etc. into a synthesized output gas stream, e.g. CO, H_2, $C_nH_mO_l$, etc.

3. Experimental setup

The atmospheric pressure microwave plasma in the MPES reactor is operated at a frequency of 2.45 GHz via a 1.2 kW magnetron (Panasonic NN-T945SF) fed by a 4 kV AC power supply. The deposited power into the plasma is in the range of 400-1000 W. The waveguide components consist of a launcher, isolator, directional coupler, stub tuner, tapered waveguide and short circuit.

Figure 2 schematically depicts the experimental setup. A 6.5 kW high-flux solar simulator from Kinoton GmbH is used as the concentrated solar radiation source. The simulator's power can be regulated to deliver 350-525 W of incident solar radiative power to the discharge tube by varying the rectifier's electrical current between 80 and 120 A. A mixture of CO_2 with Ar, the latter used for greater plasma stability, is used as the working gas. Mass flow controllers (ALICAT MC) were used to control the gas flows between 0 to 10 slpm.

An optical emission spectrometer (Avantes AvaSpec-ULS2048) is used to measure the efficiency of solar radiation absorption by the microwave plasma. A gas chromatographer (GC-2014, SHIMADZU) was used for measuring the amounts of CO, CO_2 and O_2 gases in the processes output gas stream.

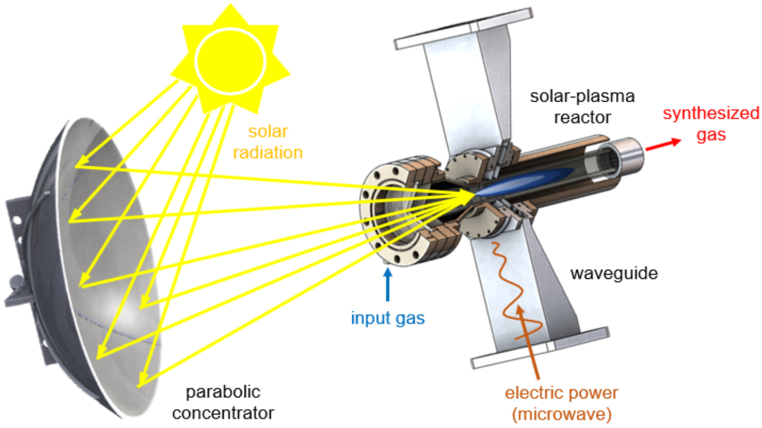

Figure 1: Microwave Plasma-Enhanced Solar (MPES) reactor design rationale.

4. MPES reactor characterization

Solar power absorption in the plasma can increase the amount of deposited power into the plasma, enhances the gas species temperature and consequently the rate of thermochemical kinetics. During the experiments, the following parameters are kept fixed: microwave deposited power (700 W), CO_2 flow rate (1 slpm), and Ar flow rate (7 slpm); whereas the solar input power is varied.

Figure 4 presents the absorption efficiency and absorbed power as function of the solar input power. The maximum solar absorption efficiency obtained is ~ 20%, occurring at the minimum value of solar input power (i.e. 350 W). The absorption efficiency decreases to ~ 14% with increasing the absorbed radiative power. The more solar input power, the lesser the fraction of solar radiation absorbed. However, the total amount of absorbed power increases from ~ 69 to 75 W with increasing the solar input power from 350 to 525 W.

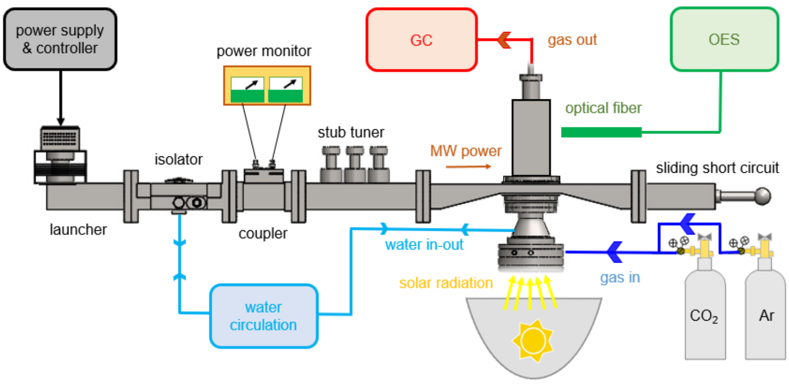

Figure 2: Schematic diagram of the MPES experimental setup.

4.1 Solar radiation absorption by microwave plasma

Figure 3 shows the optical emission spectrum of the solar radiation after it passes through the MPES reactor, with and without an established microwave plasma. The spectra were measured with the optical spectrometer probe located at the outlet of the reactor. The solar spectrum closely matches the natural sunlight spectrum [12]. The results in figure 9 were obtained with the simulator operating at 350 W of solar input power to the reactor. Radiation passes through the conical section of the reactor, converging at the plasma generation point. The radiation is then transmitted through the plasma along the discharge tube, to finally being detected by the optical probe at the outlet of the reactor. The absorption of solar radiation by the microwave plasma increases the intensity of light at the end of the discharge tube, indicating the net absorption of incident radiation by the plasma.

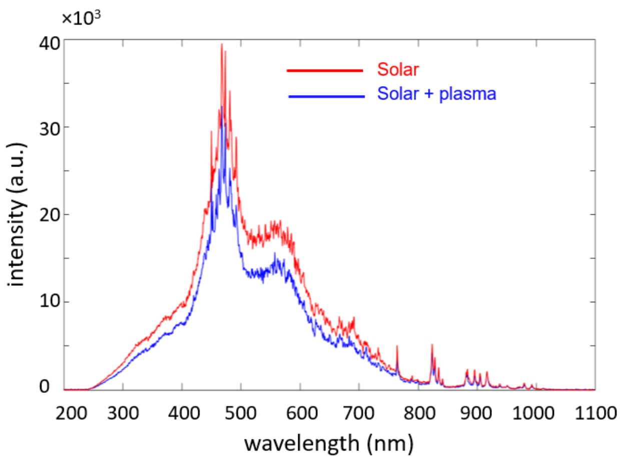

Figure 3: Spectrum of radiation exciting the MPES reactor, with and without microwave plasma.

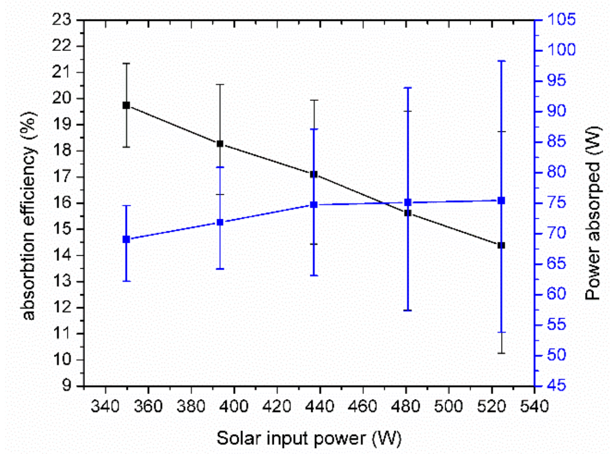

Figure 4: Solar power absorption efficiency and absorbed solar power by the microwave plasma as function of the solar input power.

4.2 Carbon dioxide decomposition via MPES

The decomposition of CO_2 by the MPES reactor was evaluated with and without solar radiation input. In these experiments, the outlet gas was cooled by a water-cooled L-shape line (as a quencher) before gas sampling.

Figure 5 shows the obtained CO_2 conversion efficiency and conversion energy efficiency. The specific electrical energy input (SEI) in all these experiments was fixed to around 1 eV/molecule. However, the input solar power changed in the range of 350-525 W. The results indicate that by introducing solar radiation into the reactor, both, conversion and conversion energy efficiency increase from 6% and 17% (when solar simulator was off) to around 9% and 25% (when the solar simulator operated at maximum power), respectively. These results are consistent with the results presented in figure 4, e.g. the solar absorption power increases with increasing solar input power. The more solar power absorbed by the plasma, the more energy deposited in the plasma that leads to greater conversion of CO_2 molecules.

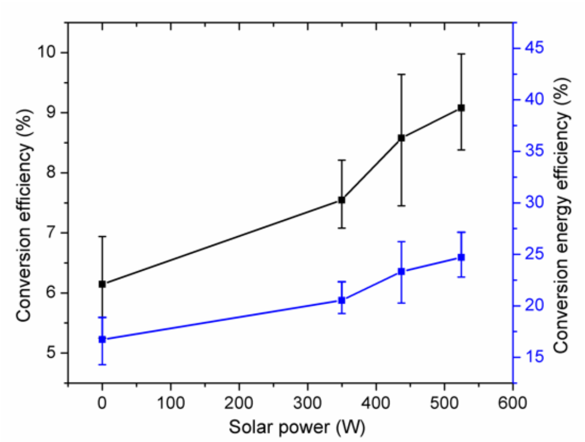

Figure 5: Conversion efficiency, plasma energy efficiency, and solar energy efficiency of CO_2 decomposition in the MPES reactor as function of input solar power.

5. Conclusion

The conversion of CO_2 using renewable energy can help mitigate environmental emission and address global fuel needs. Solar thermochemical processes have the greatest sustainability potential, but are limited by the low reactivity of CO_2 and the intermittency of solar radiation. In contrast, plasma processes have the potential for high efficiency and continuous operation. Microwave Plasma Enhanced Solar (MPES) is investigated as an approach for the synergistic combination of microwave plasma and concentrated solar radiation for CO_2 conversion processes with potentially greater viability. A MPES reactor was designed and characterized for atmospheric pressure CO_2 decomposition. Optical emission spectroscopy measurements indicated that up to 20% of incident solar radiation can be absorbed by the plasma. The solar absorbed power increases with increasing input solar power. Gas chromatography analyses showed that both conversion and conversion energy efficiency increased with increasing solar power input. This enhancement is attributed to the higher solar power absorbed by the plasma.

6. References

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