Optimization of gas quenching of atmospheric CO_2-plasma sustained in microwave reactor by application of water-cooled nozzles

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In the last decade, the increasing application of renewable energy sources, particularly based on wind and solar power, enabled the gradual replacement of fossil fuels that is necessary for the decarbonization of industry. At the same time, the inherent intermittency of these sources urges technologies to store the surplus of renewable electricity [1]. One of the most advanced technologies is the synthesis of liquid fuels, which thanks to its high energy density and extended storage time, surpasses alternatives like flywheels, batteries, or compressed air storage [2]. A particularly promising approach involves the endothermic reduction of carbon dioxide (see eq. 1) from industrial emissions, which not only serves to energy storage but can also contribute to the mitigation of greenhouse gas [3].

$$CO_2(g) \rightarrow CO(g) + 1/2\ O_2 + 283\ kJ/mol, \qquad (1)$$

The characteristic times of natural fluctuations of wind and solar energy can scale down to several minutes and to follow these fluctuations the conventional chemical reactors are often too slow. Plasma reactors allow to start/stop the gas activation process within seconds and is therefore well suited for the intermittent availability of electricity [4]. So far, plasma reactors driven by microwave energy have demonstrated the highest efficiency (over 80%) for the conversion of CO_2 to CO at low pressure conditions [5]. In general, atmospheric systems are more robust and cheaper and therefore are more appropriate for future industrial applications as compared with vacuum ones. At the same time, the efficiency of the process gets worse when the pressure increases that urges the temperature quenching in plasma afterglow. A promising approach for quenching is the application of a water-cooled metallic nozzle in the afterglow region which serves both for effective gas cross-mixing and gas cooling [6]. Very earlier experiments in 1980s with supersonic acceleration of gas in the nozzle at vacuum conditions have shown an energy efficiency up to 90% [7]. In last 5-7 years, the nozzle configurations again have drawn new attention, particularly for atmospheric CO_2 plasmas sustained with microwave [8-11]. It was shown that fast quenching with water cooled nozzle promotes both efficiency and conversion [9-11]. Despite of very promising results, the authors claim that the design of quenching system is far from optimum and there is a room for optimization [11].

In present work, we report on the optimization of gas quenching in the afterglow of atmospheric CO_2-plasma sustained in a Surfaguide microwave reactor by means of water-cooled nozzle constructions. In COMSOL Multiphysics, the flow dynamics of a gas mixture is modeled, whose effective heat capacity, heat conductivity, viscosity and density is a result of contribution of different gas species for a given temperature. The gas supplied through a helical injector flows through the reactor tube which included an effective heat source (< 1.6 kW) and further through the nozzle construction. To optimize the nozzle the taper angle (and correspondingly its length) as well as the nozzle diameter and nozzle length were varied (see Fig. 1 left). For practical reasons, two nozzles were fabricated for validation in the experiment: with a taper angle of 45° and 78° that corresponds to a difference in a gas-to-metal heat flux of a factor of 3.3. Experiments have demonstrated that the improvement through the application of nozzles compared to the no-nozzle reference case is of a factor of 4 and 2.5 for conversion and efficiency, respectively. As for the difference between two nozzle configurations, the “short” nozzle (45°) has shown some better performance as compared with the “long” (78°) nozzle (see Fig. 1 right).

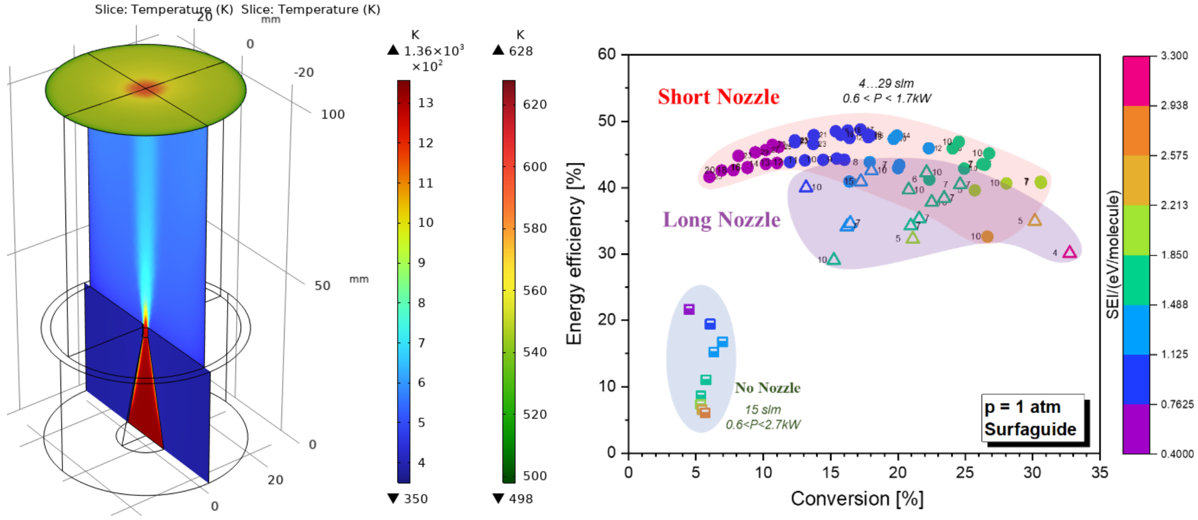

Fig. 1: (**left**) Simulated gas temperature distribution for in- and after-nozzle domains. Left scale represents the temperature in axial cut plane, and right scale corresponds to temperature in cross plane which is 70 mm away from nozzle end.
(**right**) Energy efficiency vs. conversion for no-nozzle configuration (15 slm, power scan) against the two nozzle geometries: 45° („short“) and 78° („long“) where both gas flow rate and power are varied. SEI is color coded.

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