Critical view on CO_2 conversion in warm plasmas: reactor design and life cycle assessment

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Abstract: We tested several new electrode configurations in a gliding arc plasmatron (GAP) reactor to investigate possible improvements in CO_2 conversion and energy efficiency. Although the reactor design does influence the performance, the possibilities for improvement remain limited, as the best results give only slightly higher CO_2 conversion than previous works. To investigate the possible environmental benefit of this technology, we complement these experiments with a life cycle assessment.

Keywords: CO_2 conversion, gliding arc, reactor design, life cycle assessment

1. Introduction

The current linear carbon economy leads to increasing CO_2 emissions and we need urgent action for the transition to a more sustainable society [1]. Since carbon products will not disappear entirely, careful management of the CO_2 that is already in the atmosphere and recycling human CO_2 emissions will be key to minimizing the environmental risks [2]. Carbon capture and storage (CCS) is the most promising group of technologies that can effectively decrease the CO_2 emissions before 2050 [3], but large-scale implementation is only just starting [4]. Therefore, a complementary mitigation pathway is to utilize this captured CO_2 as a feedstock for cleaner processes (carbon capture and utilization, CCU) [5]. In this context, several technologies are being developed for CO_2 conversion, including plasma technology [6].

Different types of plasma reactors have already been examined for CO_2 conversion [7] and the gliding arc plasmatron (GAP) is one of the most promising configurations [8, 9]. For pure CO_2 splitting, the GAP reactor has been previously shown to achieve energy efficiencies up to 30%, although the conversion has so far remained limited to a maximum of 8.6% [9]. In order to become a competitive technology [7] and address the scale needed for climate change mitigation [3], further improvements are needed.

Smart reactor design is essential to enhance the performance of gas conversion, as demonstrated in numerous works [10-13]. In case of the GAP reactor, the design was previously investigated by Trenchev et al. [14] with a combined modelling approach of 3D fluid dynamics and 2D plasma chemistry. Although the reverse vortex flow (RVF) is beneficial to stabilise the discharge in the centre of the reactor, it was suggested that not all the gas passes through the discharge zone, i.e. a significant amount of gas seems to leave the reactor without being in touch with the plasma. Their findings confirmed the experimental observations of Ramakers et al. [9] who found that a smaller outlet diameter of 7.08 mm yielded a much higher CO_2 conversion of 8.6% compared to 6% and 5% for the larger diameters of 14.2 mm and 17.5 mm, respectively. They attributed this improved performance to the more pronounced RVF in the design with the smallest outlet diameter and argued that this forces a longer residence time of the gas in the plasma. This stronger RVF also provides thermal insulation of the discharge from the walls, which lowers the thermal losses. In addition, the reactor with the smallest diameter displayed the longest afterglow. Such a larger active plasma volume could also explain the improved performance. Varying the outlet diameter yielded promising results, but so far, no further reactor design improvements were investigated.

In this work, we explore several new variations in electrode shapes within the existing reactor, to investigate the influence of GAP reactor design on the CO_2 conversion and energy efficiency. Moreover, to analyse the possible environmental benefit of this technology in industry, we complement our experiments with a life cycle assessment.

2. Experimental

The experimental setup is similar to the setup described by Ramakers et al. [9] Figure 1 illustrates the electrode design of this GAP, using a reverse vortex flow (RVF).

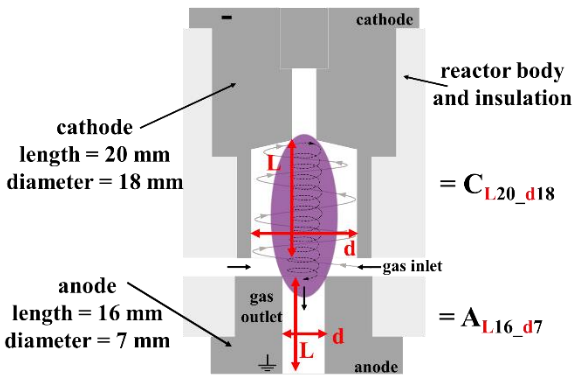

Figure 1 Schematic 2D representation of the basic GAP, with (at the right) indication of how the cathode and anode are called, based on their dimensions (see below).

The gas flows into the reactor through tangential inlets, and an arc forms between both electrodes (purple). First, the cold gas from the inlets flows upwards close to the walls (outer spiral) creating an isolating and cooling effect.

Afterwards, it flows downwards in a reverse vortex (inner spiral) where it mixes with the plasma. The electrode dimensions are indicated by red arrows (d = diameter and L = length) which is included in the name of each electrode (indicated on the right of the figure: C_{L20_d18} and A_{L16_d7}) and summarized in Table 1.

Table 1 Overview of the electrode dimensions tested

Electrode	Length (mm)	Diameter (mm)	Shape
cathode	10	18	cylindrical
	20	18	cylindrical
	30	18	cylindrical
	20	10	cylindrical
	16	18	cone
anode	16	3.5	cylindrical
	16	7	cylindrical
	16	14	cylindrical
	30	7	cylindrical
inserted anodes	30	8	cylindrical
	30	8	tapered
	30	4	tapered

The variations were limited within the same outer shape of the electrode (dark grey in figure 1) to guarantee a good fit in the surrounding reactor (light grey in figure 1), hence larger dimensions were not feasible. Smaller sizes were not possible either, since the gas volume would become too small for the flow rates of interest and the pressure would increase above safe levels.

We measured the CO_2 and CO concentrations with an online NDIR (Non-Dispersive Infrared Spectroscopy, Emerson XEPG) and the O_2 concentration with an optical oxygen sensor (Pyroscience). The CO_2 conversion and energy efficiency are determined according to the formulas described in [13]. Each experiment is repeated three times, in order to apply a propagation of uncertainty to the results and calculate the error bars.

3. Results and discussion

All the results are summarised in figure 2. The energy efficiency is plotted as a function of CO_2 conversion to give an overview of the two most important performance parameters. Both graphs display the same data, but are grouped for each anode (a) or cathode (b). The schematic representations of these electrodes are given on each side, indicated by the symbol in the graph.

Clearly, changing the electrode design has a large influence on the performance. The design with highest energy efficiency (i.e. 30%, for a conversion of 9.5%) is the combination of the longest cathode (C_{L30_d18}) with the smallest anode diameter ($A_{L16\_d3.5}$). On the other hand, the CO_2 conversion is slightly higher (i.e., 10.5%) if we combine the same (longest) cathode (C_{L30_d18}) with the

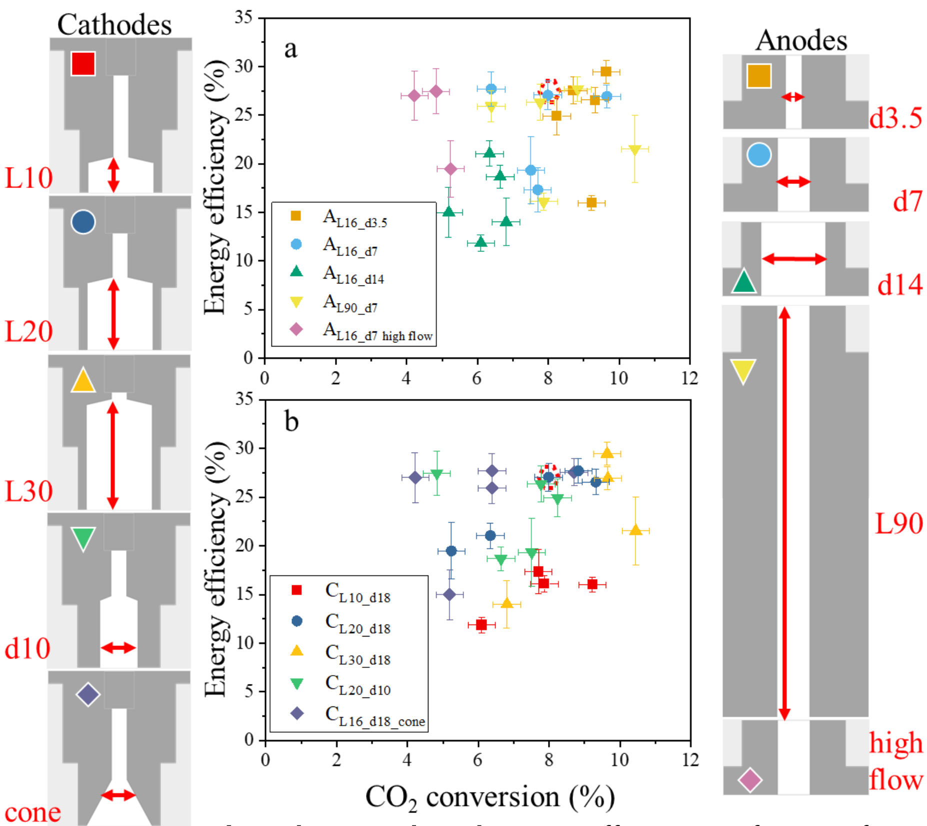

Figure 2 Performance of all electrode combinations shown by energy efficiency as a function of CO_2 conversion: (a) grouped for each anode and (b) grouped for each cathode. The schematic representations are displayed on the left for the cathodes, and on the right for the anodes. The red arrows indicate the characteristic dimension, also written next to the scheme. The red dotted circle in the graph indicates the basic combination C_{L20_d18} A_{L16_d7} from [9].

longest anode (A_{L90_d7}), but the energy efficiency here is lower (i.e., 21%), which can be attributed to the heat losses to the walls.

Some criticism is justified here: despite the large variations in performance for the different designs, there is no large improvement compared to the basic GAP design (indicated with red dotted circle), even for the best designs we tested. Many electrode designs perform worse than the basic design, which indicates that the basic GAP reactor design was already quite optimized and only varying the electrode dimensions does not lead to significant improvements. The question arises whether it would then be better to investigate a completely new design.

To answer this question, we compared our results to the performance of similar plasma types in our lab. More specifically, we considered warm plasma reactors at atmospheric pressure with contact between the electrode and the plasma: the confined atmospheric pressure glow discharge (APGD) [15], the dual vortex plasmatron (DVP) [13] and the rotating gliding arc (RGA) [16]. Despite their completely different reactor designs and operating conditions (power and flow rate), the performance of these warm plasmas is very similar, yielding a CO_2 conversion around 10% for an energy efficiency around 30%. They all seem to bump into the same limits as the GAP studied in this work, where the conversion is at maximum 10% for the "best" designs. Some conditions with higher energy efficiency are possible, e.g. at higher flow rates, but this results in such low CO_2 conversion that it is not interesting from an industrial point of view. From these comparisons, it appears that this plasma type (high temperature, pure CO_2, with contact between electrode and plasma) has a certain limit in performance, independent of the reactor design.

Instead, the performance could be enhanced by various other solutions, ranging from different plasma types (e.g. DBD [17] and low pressure MW [18]) to the design of the post-plasma zone (e.g. adding carbon as a solid reactant [19] or a nozzle for quenching [20, 21]. However, each improvement strategy has its advantages and disadvantages. Furthermore, we cannot draw any conclusions on the possible environmental benefit from these lab scale experiments. Therefore, we performed a life cycle assessment to investigate the real carbon footprint of the process on an industrial scale.

4. Life cycle assessment (LCA)

Effective sustainability tools such as Life Cycle Assessment (LCA) are necessary to monitor the environmental footprint of processes. It is a comprehensive tool for quantifying the environmental impacts of a product, process or service across its whole life cycle and can provide insights in the early stage of process design and development [22].

Our results show that a recycle loop is crucial in an industrial process to recover the unconverted CO_2 and improve the waste management. Especially the combination with bio-charcoal as a solid reactant behind the reactor [19] is interesting to decrease the production cost: it completely removes the O_2 from the mixture and reduces the energy cost of subsequent separation steps. Prioritizing such process design optimisation seems crucial to obtain a real green production technology.

5. Conclusion and outlook

Our results indicate that reactor design is less important once the CO_2 conversion reaches about 10% in this plasma type. Different plasma types and improvement strategies could be implemented, but they all have their own challenges. We performed a life cycle assessment to bridge the gap between lab scale experiments and green industrial investments. We believe that detailed process design optimisation and pilot-scale demonstrations are a crucial next step to indicate which plasma technologies are fit for specific markets. Only then can plasma technology become a valuable part of all CCU technologies that are necessary for the transition to a more sustainable world.

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