

1 Enhancing CO₂ conversion with gas quenching in arc plasma

2 Rani Vertongen¹, Ivan Tsonev,¹ and Annemie Bogaerts¹

3 ¹ Research group PLASMANT, Department of Chemistry, University of Antwerp, Universiteitsplein 1, 2610
4 Antwerp, Belgium.

5 Corresponding author: Rani Vertongen, rani.vertongen@uantwerpen.be

6

7 Abstract

8 We investigated post-plasma gas quenching in an arc plasma for CO₂ conversion. We present
9 experiments with a basic pin reactor, along with four different methods for quick cooling, i.e., a nozzle,
10 wall cooling, combining the nozzle and wall cooling, and a heat exchanger. We demonstrate that
11 quenching can significantly improve the performance. The best results are obtained with a heat
12 exchanger, enhancing the conversion with a factor three (from 6 to above 18 %) and the energy
13 efficiency with a factor 1.5 (from 20 to 30 %) when compared to the benchmark. Temperature
14 measurements indeed confirm that the heat exchanger provides the most effective cooling. In
15 addition, the heat exchanger ensures a more stable and elongated plasma which further improves the
16 performance. Interestingly, we observe a linear increase between conversion and specific energy
17 input, resulting in a constant energy efficiency of about 30 %, which is very promising for further
18 upscaling towards higher specific energy input.

19

20 Keywords

21 Plasma; CO₂ conversion; arc; quenching; reactor design

22

23 Highlights

- 24 • Significant improvement of CO₂ conversion with post-plasma gas quenching.
- 25 • Heat exchanger results in three times higher conversion (from 6 to above 18 %) than the
26 benchmark.
- 27 • Quenching ensures a constant energy efficiency of 30 % in the full SEI range.
- 28 • Stable plasma in takeover mode can elongate the arc and improve performance.
- 29 • MW and arc plasma demonstrate the same performance under similar conditions.

30 1. Introduction

31 Global warming is a complex problem and the pressure for change is high. Our greenhouse gas
32 emission must be greatly reduced, preferably to net zero, to limit the worst consequences of climate
33 change [1]. Direct electrification of industrial processes and innovative carbon capture and utilization
34 are important strategies to pursue [2]. Among the novel electrified technologies such as
35 electrochemistry, plasma reactors are gaining increasing interest for various applications, such as
36 nitrogen fixation for fertilizer synthesis and CO₂ conversion into value-added products [3–6]. Especially
37 so-called warm plasmas are promising, because of their better energy efficiency than cold plasmas.
38 Indeed, stable molecules like CO₂ are easily decomposed by the high temperature (order of several
39 1000 K) characteristic for these plasmas [3,5]. Plasma reactors also have the advantage of instant
40 process control with immediate production, thus coupling very well with fluctuating renewable
41 electricity, and they do not rely on rare metal catalysts for good performance [6].

42 Arc plasma reactors are interesting for CO₂ conversion, thanks to their operation at atmospheric
43 pressure [7]. They are characterized by relatively high gas temperatures, typically higher than 3000 K,
44 enabling fast thermal reactions [3]. An important benefit is their flexible design and easy ignition, since
45 no coupling to electromagnetic waves is needed to sustain the arc, as is the case for microwave (MW)
46 discharges. However, our previous research has indicated that the conversion in such arc reactors,
47 including gliding arcs, is limited to typically 10 % for a maximum energy efficiency of 30 % [8].

48 One of the most promising strategies for performance improvement is gas quenching, either to
49 enhance the conversion or to tune the selectivity in plasmas operating (near) thermal equilibrium, e.g.
50 in the production of acetylene from CH₄ plasma [9], and in CO₂ conversion [10]. By quickly cooling the
51 plasma afterglow, the temperature drops fast enough to protect the products from reverse reactions
52 in the effluent, e.g., for CO₂ conversion:



53 The stable products are preserved in the case of ideal quenching, while reactive atoms and radicals are
54 converted back into the initial reagents during cooling. In the case of super-ideal quenching, additional
55 conversion occurs during the cooling process, when the atoms and radicals would react with the feed
56 gas, converting it further into the products (i.e., the equilibrium of R2 shifts to the left) [11,12].

57 Several different experimental design principles have been applied already for quick cooling in CO₂
58 plasma reactors. For example, gas mixing by injecting cold gas in the hot stream can ensure high cooling
59 rates. Chekmarev et al. applied counterflow quenching with the cooled gas from the reactor, leading
60 to a factor four increase in CO₂ conversion (from 6 to 24 %), and four times increase in energy efficiency
61 (from 5 to 20 %) [13]. Placing a nozzle constriction at the outlet can have a similar effect as gas mixing,
62 since it increases the turbulence after the plasma, thereby mixing the hot core with the colder
63 surrounding gas and improving heat loss to the walls [14,15]. Hecimovic et al. applied a cooled nozzle
64 to a MW discharge, with significant improvements at 900 mbar and low CO₂ flow rates. Their CO₂
65 conversion increased with a factor seven (from 5 to 35 %) and the energy efficiency with a factor four
66 (from 5 to 20 %) [15]. Mercer et al. obtained the largest relative increase in performance at a pressure
67 of 300 mbar, with almost three times higher conversion when applying the converging-diverging nozzle
68 (from about 10 to 30 %) and three times higher energy efficiency (from 8 to 23 %). At a higher pressure
69 of 700 mbar, however, the improvement was only a factor 1.2 for both conversion and energy
70 efficiency [16]. Li et al. combined a contracting nozzle with an argon arc plasma with CO₂ addition.

71 They observed an increase in effective conversion (from 2.4 to 22 %), i.e. a factor nine higher with the
72 nozzle, and a parallel improvement in energy efficiency up to 18 % [17]. Some experiments at lower
73 pressure with nozzles additionally aim for supersonic gas expansion in the effluent, as an alternative
74 way to boost the cooling by converting the thermal energy into directional kinetic energy [15,18].

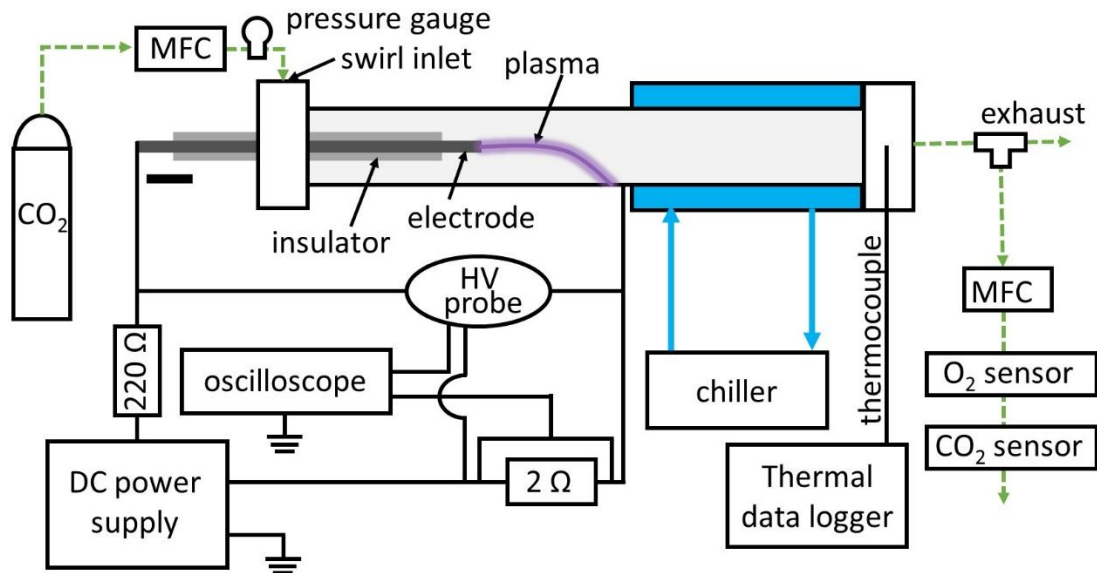
75 Cooling by direct contact with a cold wall can also improve the performance, for example by a
76 surrounding double wall [19], a cooling rod in the center of the afterglow [20], or in multiple cooled
77 outlet channels, similar to shell-and-tube heat exchangers [21,22]. Some of the best results to date
78 were achieved by Hecimovic et al. in a MW discharge at atmospheric pressure with a heat exchanger
79 (i.e., four cooled effluent channels) [22]. At a high specific energy input of $7 \text{ eV molecule}^{-1}$, the authors
80 achieved conversions up to 57 % i.e., four times higher than the maximum of 15 % in the standard
81 configuration. For the same conditions, the energy efficiency increased by a factor 10, reaching 20 %
82 with the heat exchanger compared to 2 % in the standard design. Interestingly, the results with
83 quenching at atmospheric pressure were close to the best results in the standard configuration at low
84 pressure (200 mbar) but did not exceed the energy efficiency of about 30 %.

85 Most of the above results were obtained in MW plasmas, or otherwise in argon arc discharges with
86 CO_2 addition. To the best of our knowledge, no previous research has studied an arc plasma reactor in
87 pure CO_2 with nozzle and heat exchanger. In this work, we investigate for the first time the effect of
88 quenching on the CO_2 conversion in an arc discharge with integration of either a nozzle or heat
89 exchanger. Our arc plasma reactor exhibits a very flexible design, which permits a systematic study of
90 how quenching can improve the CO_2 conversion in arc plasmas. Furthermore, we present a direct
91 comparison with the best results for CO_2 conversion in MW discharges.

92 2. Methods

93 2.1 Experimental setup

94 The experimental setup is shown in **Figure 1**.



95

96 **Figure 1** Schematic representation of the experimental setup for CO₂ conversion in an arc plasma reactor with quenching.
97 The plasma is ignited between the high voltage electrode and the surrounding stainless-steel tube.

98 A cylinder with CO₂ gas (AirLiquide, purity 99.5 %) was connected to a mass flow controller (MFC,
99 Bronkhorst EI-Flow Select type F-201AV-50K) set to flow rates from 10 to 20 l_s/min (reference
100 conditions 20 °C and 1 atm). The gas was inserted via four tangential inlets with a diameter of 1.5 mm,
101 ensuring a forward swirling flow. The inlet pressure was monitored with a pressure gauge, to ensure
102 that the pressure was the same for all the different designs (e.g., 0.45 barg at 15 l_s/min).

103 The plasma was generated between the stainless-steel pin electrode (0.8 cm diameter), surrounded
104 by a Teflon insulator, and a grounded stainless-steel tube with 1.6 cm inner diameter and 2.0 cm outer
105 diameter. Due to the grounded steel tube, the swirling flow was needed to stabilize the plasma and
106 enable operation in a large current range. When the swirling flow is not used, like in our previous work
107 with air plasma in a quartz tube and pin-to-pin configuration [23], the plasma could not be sustained
108 in this case. The electrode distance, defined as the distance from the electrode tip to the start of the
109 quenching zone, was varied between 5 and 11 cm. The quenching designs are described in more detail
110 in **Section 2.2**. When water cooling is applied, the reactor is connected to a chiller (DZ5000LS-QX, Vevor)
111 maintaining a water temperature of 20 °C.

112 The reactor was powered by a current-controlled power supply unit (PSU, Technix SR12KV-10kW) with
113 negative polarity and a ballast resistor of 220 Ω. The current signal was determined by measuring the
114 voltage drop across a 2 Ω shunt resistor, while the voltage was measured with a high voltage probe
115 (Tektronix P6015A). These electrical signals were recorded with a two-channel oscilloscope (Keysight
116 InfiniVision DSOX1102A 100 MHz). The input current was varied from 0.7 to 1 A, resulting in a plasma
117 power between 300 and 1500 W, calculated from the product of the measured voltage and current.

118 At the outlet, the gas temperature is measured using K-type thermocouples at 55 cm from the gas inlet.
119 The outlet gas after the reactor was sampled at 0.5 l_s/min with an MFC (Bronkhorst type F-200DV Low
120 dP) and sent to an optical oxygen sensor (FDO₂, Pyroscience) and an NDIR CO₂ sensor (FlowEvo,
121 SmartGas GmbH).

122 The formulas to analyze the data are taken from our previous work [24]. Every experiment is repeated
123 three times for statistical analysis, sometimes the error bars in the figures are too small to be visible.
124 The error propagation with, e.g., the MFC measurement error, is included in the calculations.

125 The conversion is calculated with (eq.1):

$$\chi = \frac{1 - y_{CO_2}^{out}}{1 + \frac{y_{CO_2}^{out}}{2}} \quad (\text{eq.1})$$

126 Where $y_{CO_2}^{out}$ is the output fraction of CO₂. This formula is valid since we only use CO₂ as an input gas.

127 The specific energy input (SEI), is defined as:

$$\text{SEI [eV molecule}^{-1}] = \frac{\text{Plasma power [kW]}}{\text{Flow rate [l}_s \text{ min}^{-1}]} \cdot \left(60 \left[\frac{\text{s}}{\text{min}} \right] \cdot 24.1 \left[\frac{\text{l}}{\text{mol}} \right] \cdot 6.24 \cdot 10^{21} \left[\frac{\text{eV}}{\text{kJ}} \right] \cdot N_A^{-1} \left[\frac{\text{mol}}{\text{molecule}} \right] \right) \quad (\text{eq.2})$$

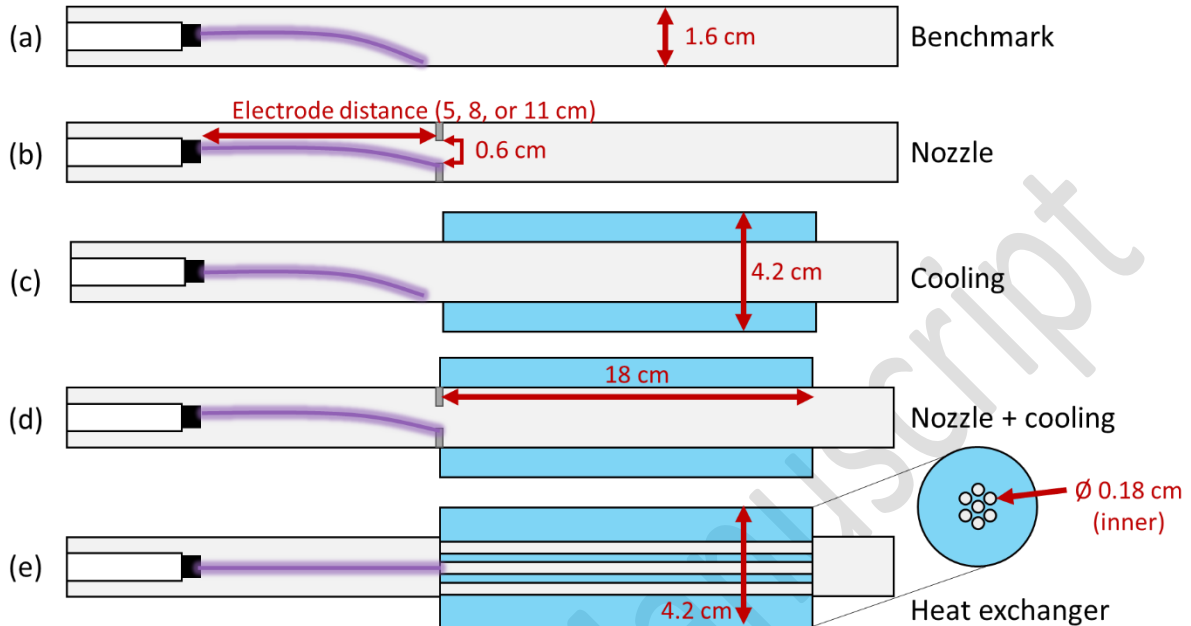
128 And the resulting energy efficiency is defined as:

$$\eta [\%] = \frac{\chi_{CO_2} [\%] \cdot \Delta H_R^\circ [\text{eV molecule}^{-1}]}{\text{SEI [eV molecule}^{-1}]} \quad (\text{eq.3})$$

129 With ΔH_R° is 2.93 eV molecule⁻¹: the standard enthalpy of the dissociation reaction of CO₂. The energy
130 efficiency is calculated based on the plasma power, which is most common in the context of plasma
131 reactor design, and it allows to compare our data with results from literature. However, energy losses
132 in the PSU or the cost of the cooling unit are not included. This would be important for calculating the
133 real impact when integrating our technology in a full process, as discussed in more detail in **Section**
134 **3.8**.

135 **2.2 Reactor designs for quenching**

136 Thanks to the simple design of the arc reactor, consisting of a pin electrode surrounded by a grounded
 137 tube counter-electrode, various quenching methods can be easily studied. We tested five basic
 138 designs, as presented in **Figure 2**.



139
 140 **Figure 2** Cross sections of the different reactor designs to investigate quenching after a CO₂ arc plasma: (a) the benchmark,
 141 (b) nozzle, (c) double wall cooling, (d) combined nozzle and wall cooling, and (e) heat exchanger designs. The important
 142 dimensions are indicated in red; when not indicated, they are the same as in the other designs.

143 The benchmark (**Figure 2a**) is a straight, stainless-steel tube with an internal diameter of 1.6 cm. The
 144 electrode tip is at a distance of 8 to 15 cm from the swirl inlet and the total length until the outlet is 50
 145 cm, similar for all designs. The nozzle (**Figure 2b**) is a round constriction with a diameter of 0.6 cm and
 146 a thickness of 1.5 cm. The cooling design (**Figure 2c**) consists of a double wall (with cooling water,
 147 connected to the chiller) with an outer diameter of 4.2 cm and a length of 18 cm. A combination of
 148 those two designs results in the nozzle + cooling design (**Figure 2d**). Finally, the heat exchanger (**Figure**
 149 **2e**) is a basic version of the shell-and-tube heat exchanger principle. It consists of seven effluent
 150 channels with an outer diameter of 0.32 cm and an inner diameter of 0.18 cm (1/8-inch Swagelok
 151 stainless steel gas lines), enclosed in a tube with 4.2 cm outer diameter. The plasma and cooling tubes
 152 are connected through flanges.

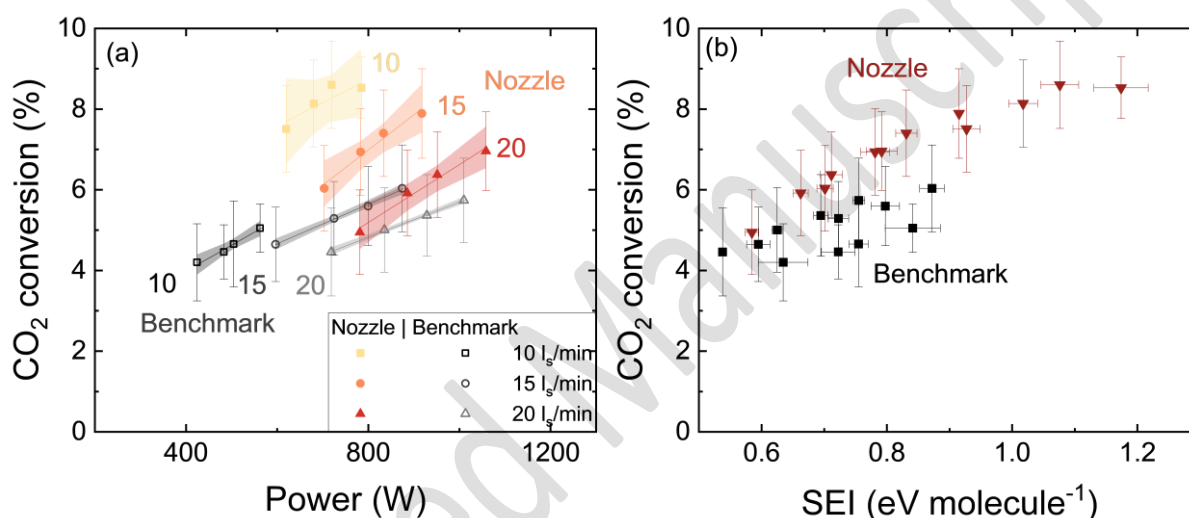
153 Although the different designs involve various restrictions to the flow in the form of a nozzle, or the
 154 heat exchanger, the effects of these on the pressure inside the plasma are minimal, as we measured
 155 the same pressure in each case. For instance, the restriction caused by the heat exchanger is minimal,
 156 due to the large number of tubes used. While the area of the nozzle is 0.28 cm², the effective area of
 157 the heat exchanger is 0.18 cm². Hence, we can safely conclude that the differences observed for the
 158 different designs are not due to different pressures.

159 3. Results and discussion

160 First, we will compare the benchmark reactor with the nozzle in **Section 3.1**. We will then present the
161 improved performance of the different cooling designs in **Section 3.2**, based on both the double wall
162 and the heat exchanger, including a discussion of the arc dynamics in **Section 3.3**. The effect of the
163 electrode distance is demonstrated in **Section 3.4**, followed by a discussion on the possible detrimental
164 effects of cooling in **Section 3.5**. **Section 3.6** gives an overview of all designs in terms of conversion and
165 energy efficiency, followed by a comparison between arc and MW designs in **Section 3.7**. Finally, we
166 list some considerations regarding the realistic application of this technology in **Section 3.8**.

167 3.1 Effect of the nozzle

168 **Figure 3** presents the CO₂ conversion at (a) different powers and (b) SEI values, for both the nozzle and
169 the benchmark. The benchmark reactor clearly shows a lower conversion than the nozzle design. The
170 energy efficiencies will be discussed in **Section 3.6**.



171
172 **Figure 3** CO₂ conversion at (a) different powers, for three different flow rates (squares for 10 l_s/min, circles for 15 l_s/min, and
173 triangles for 20 l_s/min), with an indication of the 95 % confidence interval of the fit, and (b) different specific energy inputs
174 (SEI), both for the benchmark (black squares) and nozzle (red triangles) with 11 cm electrode distance.

175 The results presented in **Figure 3a** are in line with our expectations: the conversion increases for lower
176 flow rates (corresponding to longer residence times) and higher powers. At every flow rate, the
177 subsequent points of increasing power correspond to an input current of 0.7, 0.8, 0.9 and 1 A,
178 respectively. However, the benchmark cannot couple the same power to the plasma as the nozzle,
179 even when the same conditions of input current and flow rate are applied. For example, at 10 l_s/min,
180 the power in the benchmark is at maximum 600 W, while the nozzle is able to reach 800 W at this flow
181 rate. This can be explained by the difference in plasma length between the two designs. In case of the
182 benchmark, the average voltage is 0.6 kV for all input currents at 10 l_s/min. In the nozzle design, the
183 plasma can elongate further, by attaching to the nozzle at the outlet and achieving higher voltage of
184 0.8 kV. With a higher voltage, a higher power and thus SEI (eq. 2) can be obtained. The arc dynamics
185 will be discussed in more detail in the following sections.

186 The SEI range is much larger for the nozzle (from 0.6 to 1.2 eV molecule⁻¹) than for the benchmark (0.5
187 to 0.9 eV molecule⁻¹), as indicated in **Figure 3b**. The larger SEI corresponds to a higher conversion in
188 the nozzle design, with a maximum of 8.6 %, compared to maximum 6 % in the benchmark. Considering
189 conditions with a similar SEI, the nozzle still reaches a higher conversion. At 0.85 eV molecule⁻¹ for

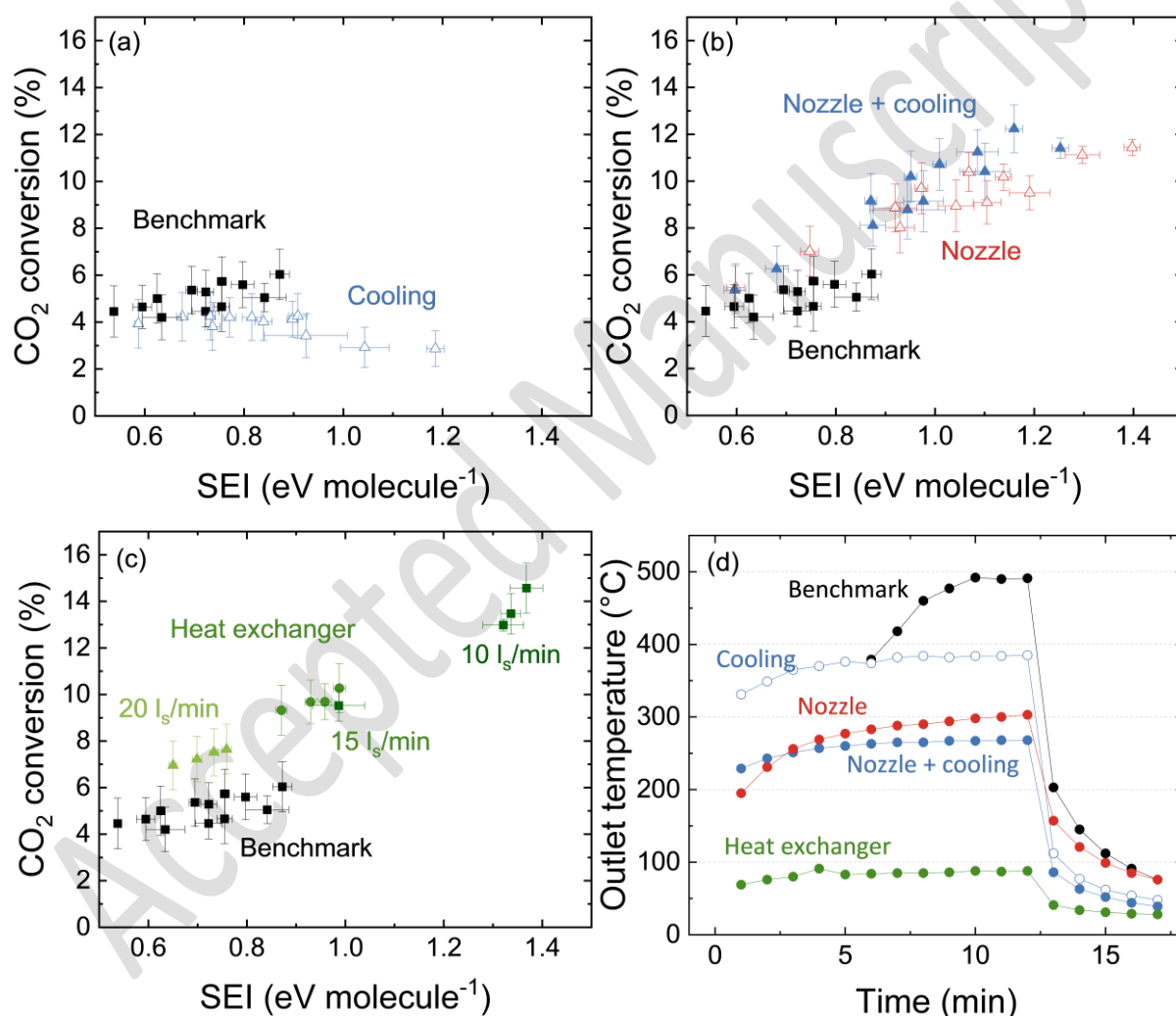
190 example, it increases from 5.0 % to 7.4 %. This indicates that, besides the increase in power, due to
191 the rise in plasma length, the nozzle has an additional effect on the performance.

192 This positive effect likely arises from the enhanced mixing after the plasma [15]. Indeed, the nozzle
193 creates turbulence and enforces the hot core gas to mix with the cooler surrounding gas, providing
194 faster cooling and limiting the recombination reactions (R1 and R2; cf. Introduction) in the effluent.

195

196 3.2 Comparing the different cooling designs

197 **Figure 4** presents the CO₂ conversion as a function of SEI, for (a) cooling, (b) nozzle + cooling, and (c)
198 heat exchanger. In (d), the outlet temperature is plotted as a function of time, to compare the cooling
199 effect of the different designs.



200

201

202 **Figure 4** CO₂ conversion as a function of SEI, for (a) cooling design (open blue triangles) compared to the benchmark (black
203 squares, (b) combined nozzle and cooling (blue triangles) compared to nozzle without cooling (open red triangles), and (c)
204 heat exchanger (green triangles for 20 l_s/min, circles for 15 l_s/min, and squares for 10 l_s/min), all with an 8 cm electrode
205 distance. In each case, the benchmark is also shown for comparison. (d) Outlet temperature as a function of time for the five
206 different designs at 20 l_s/min and 1A. The power was turned off after 13 min. Note that the benchmark was operated for a
207 shorter time, to limit the maximum temperature and protect reactor components. The measurement also started at time
208 zero, but the first data point is shifted, so that the drop in temperature aligns with the other designs.

209

210 Applying (double wall) cooling appears detrimental for the conversion, as shown in **Figure 4a**. The
211 conversion is lower than the benchmark at similar values of SEI and even decreases for higher SEI
212 values. This indicates that the onset of cooling is too early, and quenching occurs before the maximum
213 conversion is reached. This will be discussed further in **Section 3.5**. Note that a higher SEI can be reached
214 than the benchmark, even without an attachment point like a nozzle. This effect can be explained by
215 the colder gas boundary layer, increasing the drag force and pushing the attachment point of the arc
216 further downstream [25].

217 When the double wall cooling is added behind the nozzle, the conversion is clearly better than the
218 benchmark, mainly due to the higher SEI values that can be reached, similar to **Figure 3b**. However,
219 the conversion is not significantly better than the nozzle without cooling (**Figure 4b**), at least in this SEI
220 range. The nozzle + cooling design reaches a maximum conversion of 12.2 %, compared to maximum
221 11 % in case of the nozzle without cooling. It thus seems that the gas mixing due to the nozzle provides
222 more significant cooling than when the gas is in contact with the cold wall. Indeed, the nozzle has a
223 clear benefit when compared to cooling (without nozzle) (**Figure 4a**). The nozzle likely helps to maintain
224 the plasma within the reactor volume before cooling and provides the additional benefits of enhanced
225 gas mixing and plasma elongation through attachment at the nozzle.

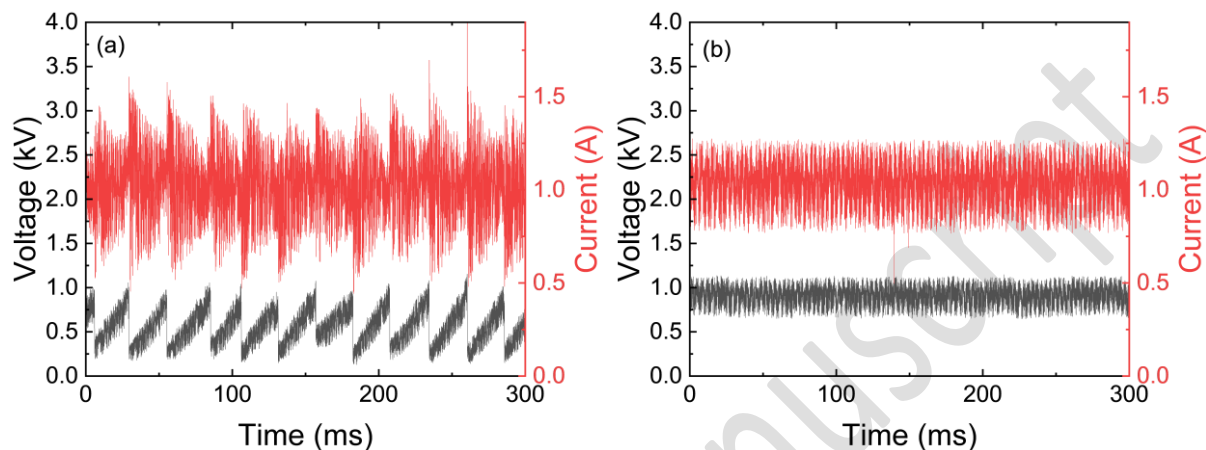
226 The cooling with heat exchanger is clearly the most beneficial. **Figure 4c** demonstrates a significantly
227 higher conversion at the same SEI values as the benchmark, for example improving from 5.7 % to 7.6 %
228 at $0.7 \text{ eV molecule}^{-1}$. Three regions can be distinguished for the heat exchanger, aligning with the three
229 different flow rates. This can be explained by the stable plasma (see **Section 3.3**) resulting in a more
230 constant power input, so that the flow rate has larger effect in resulting SEI values. The lowest flow
231 rate yields the highest SEI, and because of the (roughly) linear correlation between SEI and conversion,
232 the maximum conversion of 14.6 % is reached at the flow rate of $10 \text{ l}_s/\text{min}$, corresponding to an SEI of
233 $1.4 \text{ eV molecule}^{-1}$.

234 The temperature at the outlet in **Figure 4d** gives an indication of the different cooling capacities of all
235 designs. The outlet temperature in the benchmark case is as high as $500 \text{ }^\circ\text{C}$. Introducing (double wall)
236 cooling decreases the temperature to $385 \text{ }^\circ\text{C}$, but a nozzle is more efficient and reduces the
237 temperature to $300 \text{ }^\circ\text{C}$ without extra cooling, and to $270 \text{ }^\circ\text{C}$ in combination with cooling. This confirms
238 that gas mixing after the nozzle provides a faster and more effective way of cooling than the double
239 wall. Indeed, the improved gas mixing after the nozzle induces enough turbulence to improve the heat
240 transfer to the walls and it increases the overall heat loss in the system, explaining why the results of
241 the nozzles without and with extra cooling are so similar, as seen in **Figure 4b**. Finally, the heat
242 exchanger causes even more efficient cooling, bringing the outlet temperature down to $90 \text{ }^\circ\text{C}$. Even
243 though the surface of the heat exchanger is smaller than for the double wall cooling (58 cm^2 compared
244 to 75 cm^2), the surface to volume ratio of the heat exchanger is much larger (23 cm^{-1} compared to 2.5
245 cm^{-1}). In other words, the heat exchanger ensures a much better contact between the effluent gas and
246 the cold wall, explaining the low outlet temperature and improved performance.

247

248 3.3 Electrical characteristics of different cooling designs

249 To explain the different performance of the various cooling designs, another factor to account for is
250 the difference in SEI range between the designs. Why can the heat exchanger reach up to 1.4 eV
251 molecule⁻¹, compared to only 0.9 eV molecule⁻¹ in the benchmark? This can be explained by the plasma
252 stability, demonstrated by the voltage and current signals, as displayed in **Figure 5** for (a) the
253 benchmark and (b) the heat exchanger designs. These arc dynamics are well described for plasma
254 torches [26].



255 **Figure 5** Temporal behavior of plasma voltage and current, for (a) the benchmark and (b) the heat exchanger with an
256 electrode distance of 8 cm. Both designs operate at 10 l_s/min and 1 A input current.
257

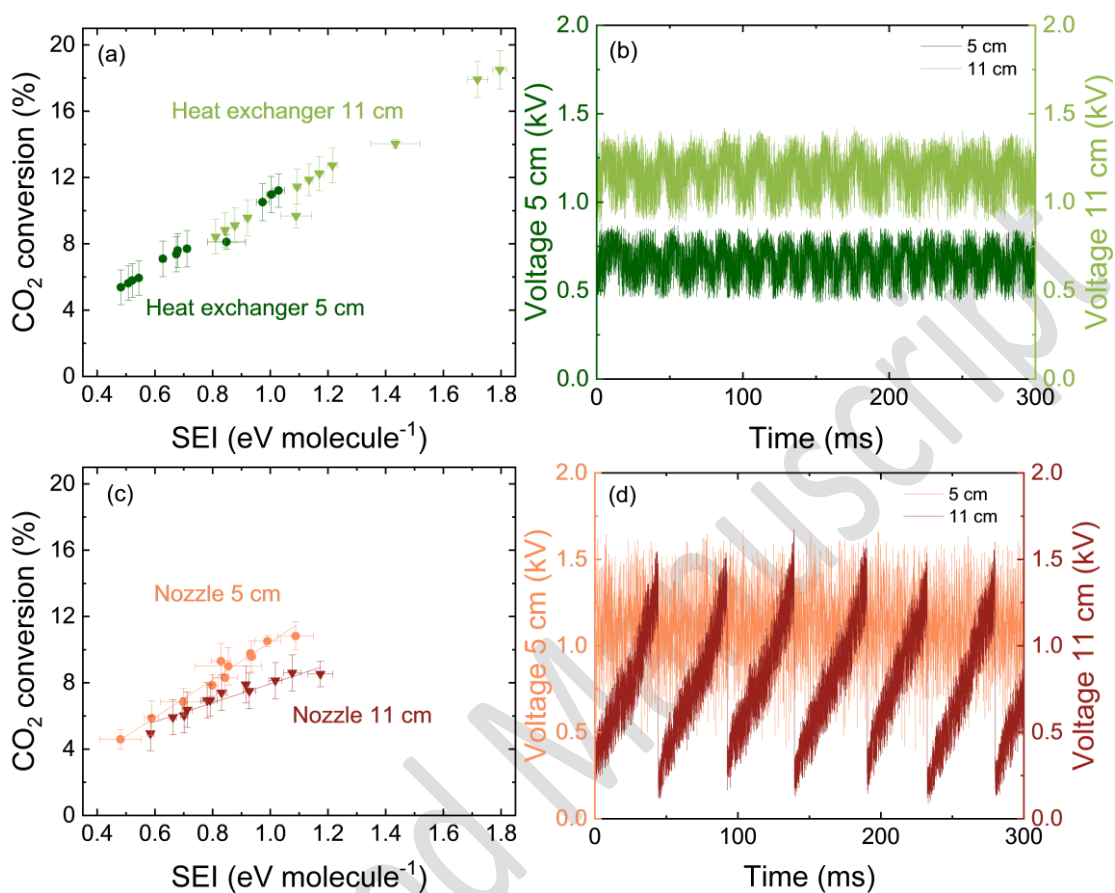
258 In case of the benchmark, the plasma exhibits the characteristic restriking mode of a gliding arc, with
259 periodic movement of the arc along the grounded electrode and typical voltage fluctuation (**Figure 5a**)
260 [27]. In the heat exchanger, however, the plasma is more stable and characterized by the takeover arc
261 regime (**Figure 5b**), probably due to a more stable attachment of the arc. In our reactor, the average
262 voltage is significantly higher in the takeover mode (e.g., 0.9 kV) than in the restriking mode (e.g., 0.5
263 kV) as seen in **Figure 5**. This yields a larger power for the same input conditions, explaining the higher
264 SEI values and thus also the higher conversion.

265 In summary, the combined effect of faster cooling and higher SEI range due to more stable plasma,
266 enhances the performance, for both the nozzle and the heat exchanger, with the latter demonstrating
267 superior performance.

268

269 **3.4 Effect of electrode distance**

270 **Figure 6** illustrates the effect of the electrode distance on the CO₂ conversion as a function of the SEI,
 271 for (a) the heat exchanger and (c) the nozzle without cooling. The temporal variation of voltage is
 272 presented in (b) and (d) for the respective designs.



273
 274
 275 **Figure 6** CO₂ conversion as a function of SEI, for two different electrode distance (circles for 5 cm, triangles for 11 cm), for (a)
 276 the heat exchanger and (c) the nozzle. Temporal behavior of the voltage for (b) the heat exchanger at conditions of maximum
 277 SEI, i.e. 10 l_s/min and 1 A; and (d) the nozzle at conditions of similar SEI, i.e. 1.08 eV molecule⁻¹.

278 The electrode distance proves to be a determining parameter for the performance. A larger electrode
 279 distance is clearly beneficial in case of the heat exchanger (**Figure 6a**). The SEI range in the 5 cm case
 280 is maximum 1.1 eV molecule⁻¹, although the rising trend in conversion is evident. With a longer
 281 electrode distance of 11 cm, the SEI can reach up to 1.8 eV molecule⁻¹ at the same conditions of 10
 282 l_s/min and 1 A. This trend can again be attributed to the longer plasma length. The plasma can attach
 283 easily to the heat exchanger and form a stable channel. A longer electrode distance thus yields a longer
 284 plasma, resulting in a higher voltage, as shown in **Figure 6b**, hence, more power can be coupled. As a
 285 result, the heat exchanger with 11 cm electrode distance can achieve much higher conversion, i.e., at
 286 maximum 18.5 %.

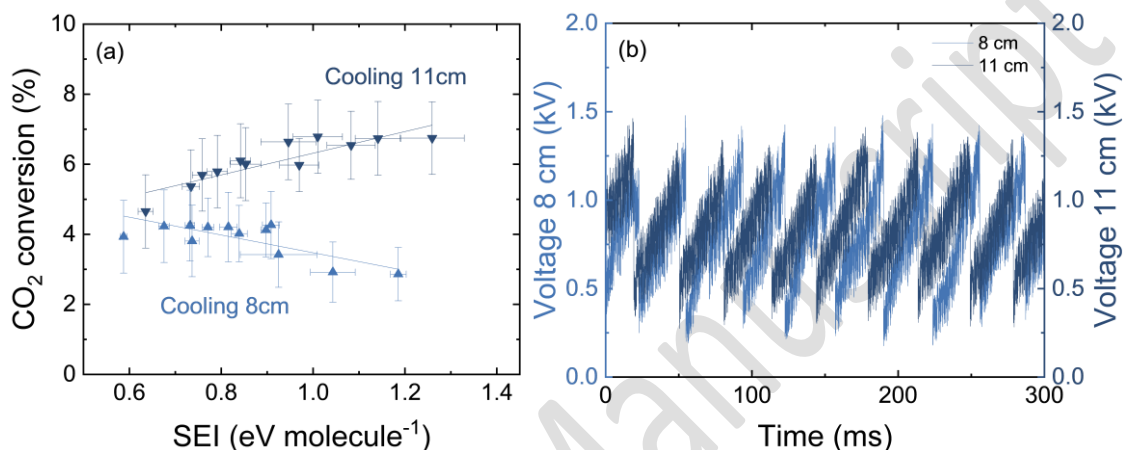
287 However, a longer electrode distance is only beneficial when comparing similar plasma modes, as
 288 demonstrated by the results of the nozzle in **Figure 6c**. The results of the shorter distance (5 cm) are
 289 slightly better than for 11 cm at similar SEI values. The voltage signal in **Figure 6d** demonstrates a clear
 290 difference between both cases. At 5 cm, the plasma operates in the takeover mode, compared to the
 291 restriking mode in case of 11 cm. The nozzle is probably more difficult to attach than the heat
 292 exchanger, due to the higher velocity in the nozzle throat [14]. Probably, the arc keeps gliding
 293 somewhere in the tube when the distance is too great, resulting in a different operating mode at 11

294 cm than at 5 cm. Consequently, the afterglow is further away from the nozzle and quenching happens
295 too late, so that the recombination reactions have taken place already, thus explaining the lower
296 conversion. In summary, these results indicate that the performance can be optimized at the largest
297 electrode distance that can maintain a stable plasma in the takeover mode.

298

299 3.5 Why does cooling not always help?

300 **Figure 7** presents the results for the cooling design at electrode distances of 8 and 11 cm, illustrating
301 (a) the CO₂ conversion as a function of SEI, and (b) the temporal voltage signal. The data at 8 cm is also
302 shown in **Figure 4a**.



303

304 **Figure 7** (a) CO₂ conversion as a function of SEI, and (b) temporal behavior of the plasma voltage for different electrode
305 distances (up light-blue triangles for 8 cm, down dark-blue triangles for 11 cm) in the cooling design without nozzle.

306 Obviously, the design with double wall cooling displays opposite trends in conversion as a function of
307 SEI for different electrode distances. The results at low SEI (~ 0.6 eV molecule⁻¹) are relatively close,
308 with a conversion between 4 and 5 % (**Figure 7a**). The conversion increases with SEI in the case of 11
309 cm, as expected, but the opposite trend is observed for the 8 cm electrode distance. The conversion
310 drops to values as low as 2.9 %, compared to 6.7 % conversion in the case of 11 cm. The difference in
311 performance cannot be explained by a difference in the plasma operation mode, as both operate in
312 the restriking mode (**Figure 7b**).

313 Instead, the observations in **Figure 7a** can be explained by the effect of quenching location. For a higher
314 SEI input, the plasma extends freely and propagates into the double wall cooler. In case of a shorter
315 electrode distance, the early onset of cooling is detrimental for the performance, because the
316 conversion has not reached its maximum value yet. This is in line with a recent combined modelling
317 and experimental study, where Ceulemans et al. [28] studied the balance between CO₂ splitting and
318 recombination reactions and demonstrated that the latter only become dominant in the afterglow of
319 a gliding arc plasma. In our cooling design with a longer electrode distance, the quenching only starts
320 after the CO₂ conversion has reached its maximum, and thus it is beneficial for the performance. This
321 negative trend for smaller electrode distances is not observed in the designs with a nozzle or heat
322 exchanger, because they have physical borders that limit the plasma size. A model specific for these
323 experiments can help to explain these trends in quenching location and will be part of future work.
324 Overall, these results confirm that the cooling needs to happen only in the afterglow for an improved
325 performance and not within the arc length for the range of SEI under study.

3.6 Overview of the performance for the different designs

Figure 8 compares the benchmark to three cooling designs in terms of (a) CO₂ conversion and (b) energy efficiency. The (double wall) cooling is not presented, because we demonstrated it typically exhibits lower performance.

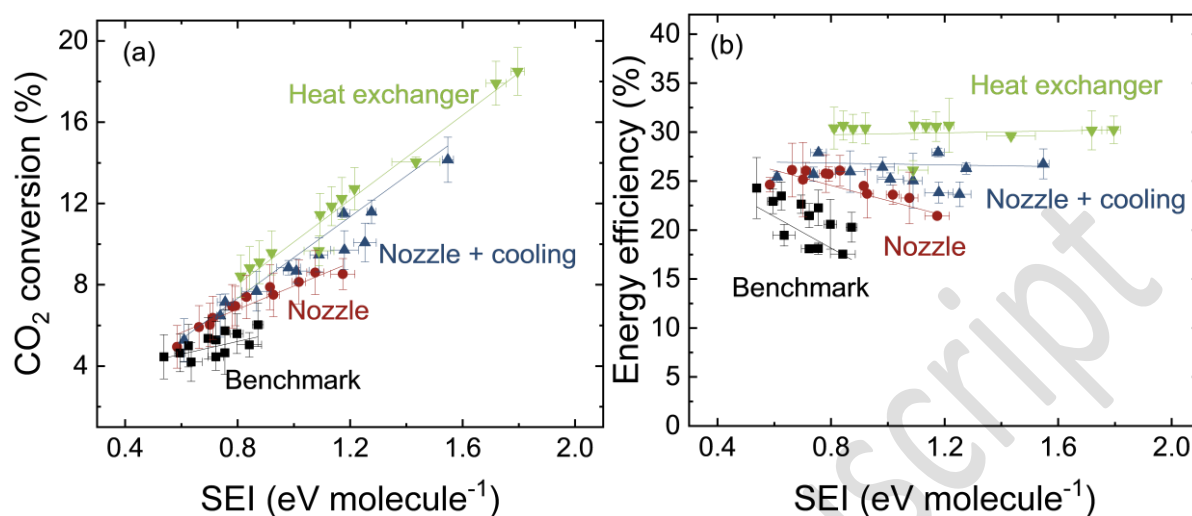


Figure 8 Overview of the results in terms of (a) CO₂ conversion and (b) energy efficiency as a function of SEI. Four designs are compared: the benchmark (black squares), nozzle without (red circles) and with cooling (blue up triangles), and heat exchanger (green down triangles). The electrode distance is 11 cm in all designs.

The enhanced CO₂ conversion is clearly demonstrated for all three cooling designs in Figure 8a. Any method of quenching will result in a higher conversion at the same SEI as the benchmark. More importantly, the quenching designs can change the plasma mode, due to the attachment of the arc to the nozzle or heat exchanger, thereby extending the plasma length. Hence, the resulting power in the plasma can be much higher, expanding the SEI range, even up to 1.8 eV molecule⁻¹ for the heat exchanger operated at 10 l_s/min and 1 A, in the stable takeover mode. The CO₂ conversion increases linearly with SEI for all designs, but the results of the heat exchanger are consistently higher than for the other designs, indicating that it is the most effective way of cooling to prevent the recombination reactions.

The energy efficiency completes our understanding of the comparison between the cooling designs, and is displayed in Figure 8b. The benchmark shows the typical trend that a higher conversion at higher SEIs is accompanied by a drop in energy efficiency. This happens when the conversion rises to a lower extent than the SEI (cf. equation 3 in Section 2.1). The nozzle also shows a downward trend in efficiency, but it is still higher than the benchmark, e.g. at 0.8 eV molecule⁻¹, the energy efficiency increases from 20.6 % to 26.1 %. Adding a cooled double wall after the nozzle yields a slight improvement in conversion, but the effect on the energy efficiency is more significant, because the trend is less negative. Finally, the heat exchanger outperforms all other designs, with a constant energy efficiency of 30 % in the full SEI range.

From our experiments, it is difficult to separate whether specific plasma-chemical interactions or flow dynamics could also play a role. Overall, our experiments suggest that simply the difference in cooling efficiency is the most important, since the heat exchanger has an outlet temperature of less than 100 °C compared to about 250 °C in the designs with the nozzles and about 500 °C in the benchmark. A model specific for these experiments, as well as sophisticated laser diagnostics, can help to explain the observed trends and will be part of future work.

358 In summary, the heat exchanger yields a factor three enhancement in the CO₂ conversion, from
359 maximum 6.0 % in the benchmark to 18.5 %, and at the same time, the energy efficiency also improves
360 by a factor 1.5, from 20.5 % in the benchmark, to 30.2 %. This clearly demonstrates that the heat
361 exchanger is the most effective cooling method in our study. Although it was previously believed that
362 extremely high quenching rates are needed to preserve products from conversion [10], our study
363 demonstrates that even a simple heat exchanger (hence, without much extra cost) could suffice to
364 mitigate the issue with recombination reactions.

365

366 **3.7 Comparison between arc and MW plasma for CO₂ conversion**

367 Interestingly, our results are comparable to the MW results reported by Hecimovic et al. [22]. They
368 were able to couple much higher powers in the plasma (up to 3 kW), corresponding to an SEI of
369 maximum 7 eV molecule⁻¹, which explains why their conversion is significantly higher (up to 60 %).
370 However, in the same SEI range up to 2 eV molecule⁻¹ and at atmospheric pressure, our heat exchanger
371 results align exactly with the results in the MW plasma, as the authors reported a conversion of about
372 16 % for an energy efficiency of 25 %.

373 Earlier works attributed the good performance of warm plasmas, such as (gliding) arc and MW plasmas,
374 to specific plasma effects (e.g., electron impact reactions and vibrational-translational non-
375 equilibrium) [6,11]. However, recent *in-situ* experiments in MW plasmas have demonstrated that the
376 heavy particles in the plasma are in thermal equilibrium with the gas [29–31]. This means that both
377 the conversion and energy efficiency have a theoretical limit that can be determined by
378 thermodynamic equilibrium calculations, as explained by Bekerom et al. [29]. At low SEI, the
379 conversion is limited when the available energy cannot dissociate all molecules. At high SEI, the
380 efficiency is limited when the energy input exceeds the reaction enthalpy for complete dissociation.
381 D’Isa et al. [31] measured a maximum energy efficiency of 30 % and a gas temperature of 6000 K, which
382 agrees exactly with this thermal equilibrium efficiency limit.

383 In our experiments, we achieved the same maximum efficiency of 30 %. Furthermore, a similar gas
384 temperature of 6000 K was measured in a pin reactor by Becarra et al. [32], indicating that the same
385 effects of thermal equilibrium are dominant, independent of the different physics that govern the MW
386 and arc plasma. This is quite striking, because the latter is heated by DC current, and the former by
387 electromagnetic fields, but it illustrates that these underlying mechanisms do not significantly alter the
388 performance. Indeed, under these conditions, the thermal chemistry is dominant, and quenching is
389 therefore essential to maintain the high conversion from the hot plasma core in the effluent.

390 Our arc plasma can probably be further optimized to achieve the same high conversions as in the MW
391 plasma, when operating at higher power (and thus, SEI), although the advantage of the latter remains
392 that they operate without electrodes and thus avoid problems of electrode erosion. For both plasma
393 reactors, however, the total efficiency of the system is essential when considering practical
394 applications. As highlighted in a recent study by Kiefer et al. [33], the efficiency of MW power supplies
395 (i.e., fraction of power delivered by the PSU that is effectively delivered to the plasma) is limited to the
396 order of 70 % at 2.45 GHz [34], while arc power supplies typically have higher efficiency (order of 80-
397 90%) [35]. Moreover, arc plasma reactors are easier to engineer, a significant advantage when
398 considering that any technology for electrification of the chemical industry has to be coupled with heat
399 integration. Until now, there are no studies showing heat recovery of the residual energy for CO₂
400 conversion through preheating the input gas. Furthermore, the residual heat could be used to activate
401 the reverse Boudouard reaction in a post-plasma carbon bed [36,37], which can further enhance the
402 conversion and energy efficiency.

403 In summary, our results show that with efficient quenching, the CO₂ conversion rises linearly with the
404 SEI, resulting in better performance. As a result, we obtain a constant energy efficiency, which does
405 not drop upon rising SEI. However, the maximum energy efficiency obtained in our work is 30 %, in
406 line with the thermal efficiency reported in literature [8,12,31]. Therefore, in our future work, we plan
407 to apply heat recovery, by using the heat removed with the heat exchanger to preheat the input gas,
408 so that the applied power can all be used for the conversion and does not have to be (partly) used for
409 gas heating. We expect that the overall energy efficiency of the system will in this way increase further,
410 important for industrial application.

411

412 **3.8 Considerations for realistic application**

413 With basic design principles, we already demonstrated significant improved performance in our arc
414 reactor. Of course, the simple setup leaves room for further improvement and some factors must be
415 considered for a more realistic application.

416 First, both the materials and geometry of the heat exchanger could be improved based on the well-
417 established heat exchanger technology in the chemical industry. The heat transfer could be improved
418 by using specific copper and nickel alloys, or even ceramics instead of stainless steel, although there
419 will be a trade-off between the increased material cost and improved performance [38]. The material
420 must also be able to withstand high temperatures, especially at the arc attachment point (above 6000
421 K), since it is crucial that the quenching happens close to the plasma. For the geometry, the surface
422 area could be increased, and extra strategies such as tape inserts or surface roughness [39] might
423 further improve the performance. From a process point of view, the heat exchanger can also facilitate
424 heat integration with other processes, which is up to now not investigated in plasma reactors.

425 Second, we should note that the energy efficiency here is calculated based on the plasma power. When
426 optimizing the reactor and PSU for realistic application, the plug power is more important. In our
427 experiments, we measured the plug power for the condition of the highest conversion, i.e., of 10 l_s/min
428 and 1 A. While the plasma power was 1.16 kW, the plug power was 2.08 kW, hence the plasma
429 accounted for 55 % of the total consumption. Accounting for the plug power in the SEI calculation, this
430 means that the energy efficiency of 30 % (based on plasma power) would decrease to 18 %, when
431 based on plug power. As mentioned in **Section 3.7**, the arc power supply can certainly be optimized
432 for a fixed reactor configuration and plasma power. For example, as explored in previous work from
433 our group [40,41], the ballast resistor could be removed if inductive elements are used or if the
434 topology of the PSU changes so that the current limitation is provided by the transformer. In this case,
435 the energy efficiency based on the plug power will be close to the plasma energy efficiency.

436 Another important factor is the long-term stability of the plasma reactor. Kiefer et al. demonstrated
437 for a MW plasma reactor that the performance remains stable for at least 30 h [33]. Similar long-term
438 stability is expected in our arc reactor, and preliminary tests of 6 h for another (gliding) arc reactor in
439 our lab revealed a stable conversion and energy efficiency within 3 %, although the electrode erosion
440 must be considered on even longer time scales. The stainless-steel pin cathode used in our
441 experiments could be further improved with better materials (e.g. tungsten alloys or graphite) or
442 protected by active cooling, i.e., strategies from commercial (larger scale) thermal plasma arc torch
443 applications [42]. Furthermore, operating the plasma reactors in parallel [35] or at lower current and
444 higher voltage will also significantly limit the erosion, as the latter is primarily determined by the
445 current. Higher currents will be needed when upscaling, but this can be optimized depending on the
446 cost of electrode erosion. On the other hand, thermal spray torches go up to 400 A, compared to only

447 1 A in our experiments, which demonstrates that there exist solutions to mitigate electrode erosion
448 and reactor stability.

449 It is important to put our results in the context of other CO₂ conversion technologies. Plasma
450 technology certainly has interesting advantages, as outlined in the Introduction and the extensive
451 review by Snoeckx and Bogaerts [6]. However, a quantitative comparison is often challenging since
452 the evaluation parameters are very different between the various technologies. Therefore, a
453 discussion on the economic feasibility and environmental impact could provide a more relevant
454 comparison. We recently performed a detailed techno-economic and sustainability analysis of a
455 scaled-up plasma process for CO₂ conversion, based on a warm plasma setup with similar parameters
456 as the experiments in this work, and we compared the metrics to electrolysis (i.e., a zero-gap type low-
457 temperature electrolyser). Both the techno-economic [43] and sustainability [44] assessment revealed
458 favourable results for the plasma process, thanks to the simple setup and cost-effective materials.

459 Specifically, the production cost of CO was estimated at \$ 671 per tonne of CO, compared to \$ 962 for
460 electrolysis, which are both competitive compared to CO transported in gas cylinders (up to \$ 3000 per
461 tonne). The electricity costs had the most significant contribution for both technologies but are
462 expected to decrease from renewable sources in the future. A sensitivity analysis also revealed an
463 optimal scenario with low-cost feedstock and equipment, so that the CO production could fall below
464 \$ 500 per tonne of CO, which is more feasible in the plasma reactors thanks to their simpler design and
465 absence of costly catalysts.

466 In terms of environmental impacts, the plasma demonstrates reductions in 7 of the 10 environmental
467 impact categories evaluated, when compared to the equivalent conventional process of partial
468 combustion with fossil fuels. The plasma process could also achieve 40 % energy savings compared to
469 electrolysis. Furthermore, adding a recycling loop of unreacted CO₂ increases the material circularity
470 indicator to above 0.8, which is 10 % higher than electrolysis. Finally, also the Green Chemistry metrics
471 are more favourable than electrolysis by around 10 – 30 %. More details can be found in our previous
472 work [43,44].

473 The upscaling of the arc plasma under study here will probably be slightly different from the case study
474 of [43,44], which was based on many plasma reactors in parallel, while the arc plasma under study
475 here should be upscaled towards industrial thermal plasma arc torch designs [42]. However, the
476 characteristics of the process will be similar.

477 In future work, such comprehensive cost and sustainability studies will also be interesting to
478 investigate for the arc plasma in this work. For example, the impact of the CO₂ source and possible
479 purification costs, as well as heat integration are interesting subjects for such studies. Since our simple
480 heat exchanger was already quite effective to enhance the performance, a passive cooling system with
481 heat integration might be sufficient for improved performance, while avoiding the energy cost of the
482 cooler. It should be noted that studies at such low technology readiness level are not applicable to
483 specific cases for companies, but modelling such costs remains a valuable tool to identify the most
484 relevant optimization strategies.

485 Overall, the findings in our study are a promising next step to improve carbon capture, utilization, and
486 storage (CCUS) technologies for the electrified production of chemicals. The rapid reduction of CO₂
487 emission is crucial to limit climate change, and decarbonization will remain the priority. However, in
488 the transition period, CCUS is essential as one of the key mitigation strategies to achieve net zero by
489 2050 [2]. Together with direct electrification, avoided demand and more renewable energy, improving
490 CO₂ conversion by plasma technology can help to achieve our climate goals.

491 **4 Conclusion**

492 We performed a systematic investigation on the effect of various quenching methods on the CO₂
493 conversion and energy efficiency in an arc discharge. We demonstrate that post-plasma cooling can
494 significantly enhance the performance by preventing the recombination reactions. Our results clearly
495 show that a stainless-steel heat exchanger provides the most effective quenching. The conversion
496 reaches up to 18 %, which is a factor three higher than the maximum of 6 % in the benchmark (without
497 quenching). In addition, thanks to the linear correlation between conversion and SEI, the energy
498 efficiency is constant upon rising SEI, reaching 30 %, a factor 1.5 higher than the maximum energy
499 efficiency in the benchmark. Introducing a nozzle in the reactor after the plasma region, also improves
500 the performance, even without wall cooling. The conversion then reaches 12 %, a factor two higher
501 than the benchmark. These results follow the same trend as reported by Hecimovic et. al. [22],
502 indicating that there is no fundamental difference in the underlying processes between a MW and arc
503 discharge and that both are governed by the thermal efficiency.

504 Furthermore, our results show that a stable arc plasma provides better conversion and energy
505 efficiency than when the arc is in an unstable regime. The unstable restriking mode is common for the
506 benchmark design, without cooling options. Introducing a nozzle or heat exchanger provides an
507 attachment point for the arc, which induces a transition from the restriking mode to the more stable
508 takeover mode. Thanks to this stable arc elongation, the average voltage is higher, and more power
509 can be coupled into the plasma, resulting in improved performance.

510 Interestingly, we show that cooling not always helps to improve the performance. We observe a drop
511 in conversion by a factor of 2.3 for an 8 cm electrode distance when compared to the 11 cm distance,
512 in the case of the double wall cooler without nozzle at SEI higher than 0.9 eV molecule⁻¹. This highlights
513 the importance of the quenching location for improved performance, i.e., cooling should not happen
514 within the arc length for the investigated SEI range, but in the afterglow, after the maximum conversion
515 is reached, and recombination reactions become dominant over the CO₂ splitting reactions. A model
516 specific for these experiments, as well as sophisticated laser diagnostics can help to explain the
517 observed trends and will be part of future work.

518 Finally, we outlined some considerations for a more realistic application of this technology. The basic
519 design of the heat exchanger already gives promising results, but the geometry and material choice
520 can certainly be optimized. Strategies to mitigate electrode erosion, as well as long term stability tests,
521 and a more comprehensive energy cost analysis with heat integration are other important steps that
522 will be part of future work.

523

524 **5 Acknowledgments**

525 We acknowledge financial support from the Fund for Scientific Research (FWO) Flanders (Grant ID
526 110221N) and the European Research Council (ERC) under the European Union's Horizon 2020
527 research and innovation program (grant agreement No 810182 - SCOPE ERC Synergy project).

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