Upscaling plasma-based CO_2 conversion:

Case study of a multi-reactor gliding arc plasmatron

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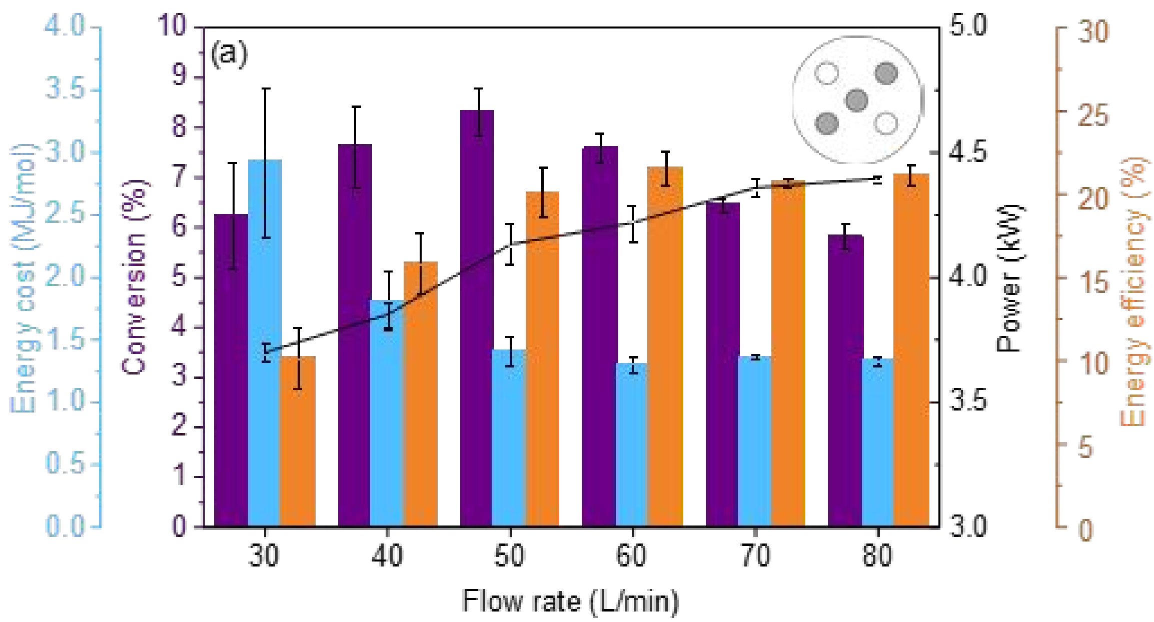
(a)
Energy cost (MJ/mol)
Conversion (%)
Power (kW)
Energy efficiency (%)
Flow rate (L/min)
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
0
1
2
3
4
5
6
7
8
9
10
3.0
3.5
4.0
4.5
5.0
0
5
10
15
20
25
30
30
40
50
60
70
80

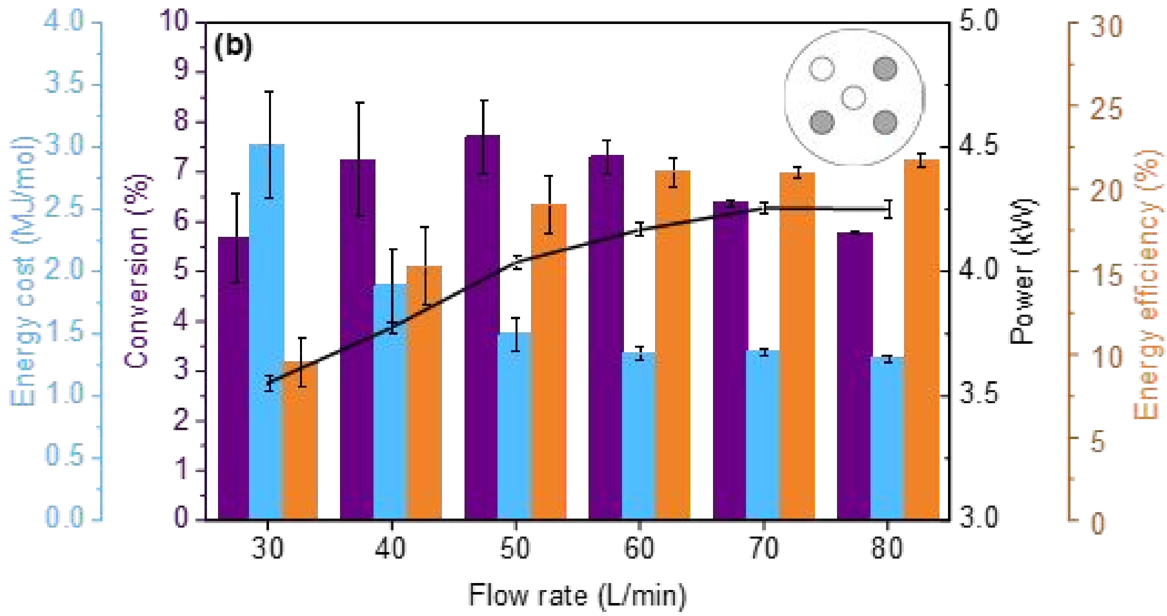

Figure S1. Energy cost, conversion, power and energy efficiency of three active reactors positioned linearly **(a)**, and three active outer reactors **(b)** (shown inset) as a function of the feed gas flow rate.

With three active reactors (Fig. S1a and S2b), the conversion profile as a function of increasing flow rate differs from that observed with one active reactor. The conversion initially rises from ca. 5.5% at 30 L/min, to a maximum value around 8% at 50 L/min. At higher flow rates (60-80 L/min), the conversion decreases again, returning to a value around 5.8% at 80 L/min in both configurations (similar to the values obtained at 30 L/min). Despite this peak-like behaviour in conversion, the energy efficiency of the process continually increases as a function of flow rate, rising from 9.5–10% at 30 L/min to a maximum value of 21% at 60 L/min. At higher flow rates, the energy efficiency remains relatively constant around this maximum value. As the flow rate increases, the power in both configurations continually rises, increasing from 3.6-3.7 kW at 30 L/min to a maximum value around 4.3–4.4 kW at 80 L/min. The energy cost of the entire process also shows a positive trend with the upscaled results, decreasing as a function of flow rate from 2.9 – 3 MJ/mol at 30 L/min to a minimum value around 1.3 MJ/mol at 50 L/min, remaining around this lowest value at higher flow rates in both configurations. At 30 L/min, the low conversion is likely due to the fact that the post-plasma chamber temperature is slightly too high (ca. 460 ˚C measured at the temperature probe T2), facilitating more back-reactions, i.e., oxidation of CO to form CO_2. It should be noted that the temperature measurements taken correspond to the bulk gas temperature, while the temperature within the plasma will in fact be much higher (up to an order of magnitude).

At 80 L/min, the fraction of gas treated by the plasma is the lowest, similar to the single reactor configurations at high flow rates. This leads to low conversion values, but the highest energy efficiency, likely because the re-oxidation is minimal, and all product (CO) remains.

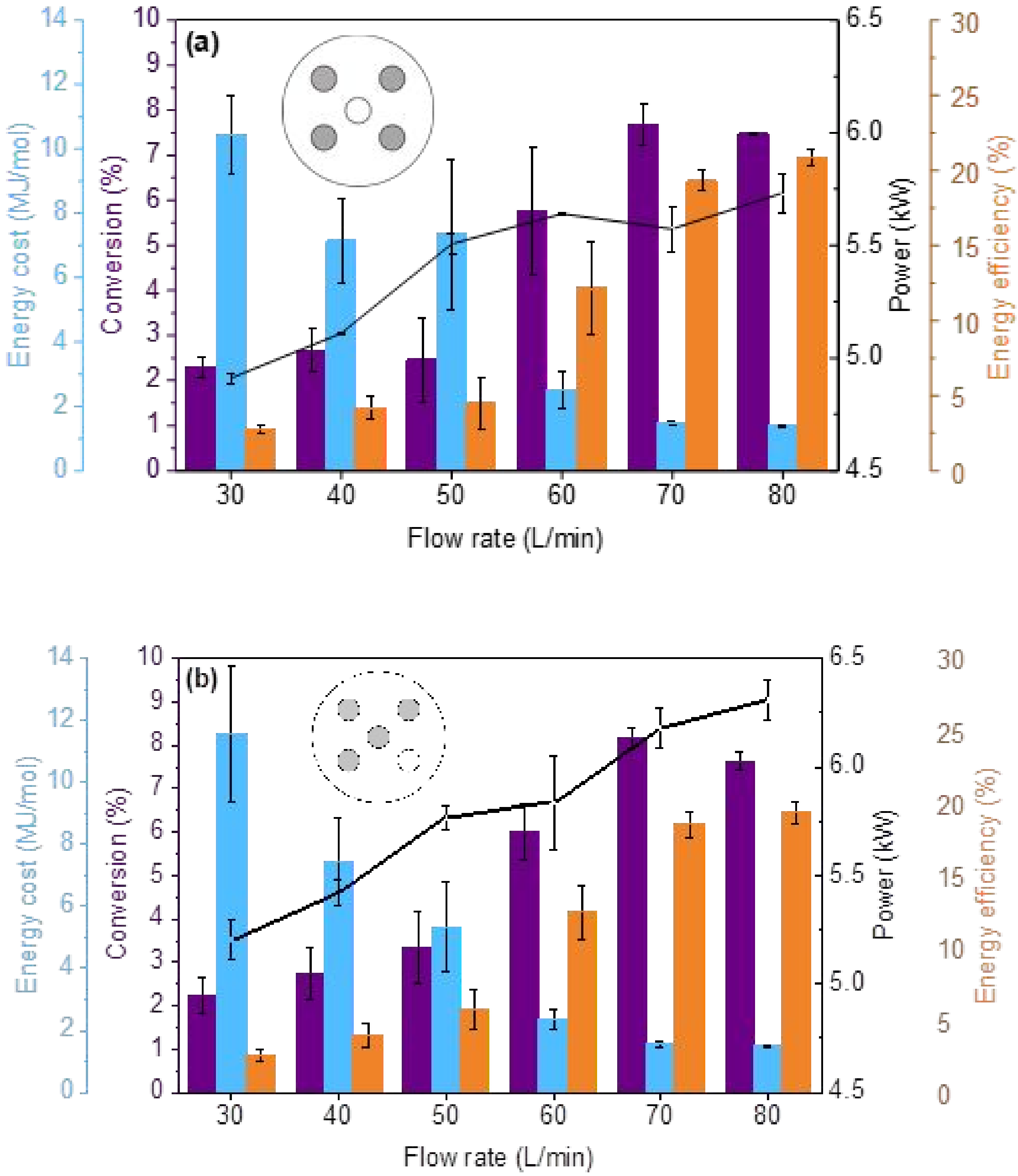

Figure S2. Energy cost, conversion, power and energy efficiency of four active reactors as a function of the feed gas flow rate, for configurations with four active reactors separated and equidistant from each other **(a)** and condensed **(b)**.

The peak behaviour observed with three active reactors is also present in both configurations with four active reactors (Fig. S2a and S2b). In general, we observe a trend of the peak performance shifting towards higher flow rates with the use of more active reactors. This is especially important for the purpose of upscaling using this principle of parallelisation of plasmatrons.

The conversion increases from 2.3% at 30 L/min to a peak value between 7.7 and 8.2% at 70 L/min. At an even higher flow rate (80 L/min), this value begins to decrease again. The energy efficiency follows the same trend as with fewer active reactors, rising from a low value around 3% at 30 L/min to its maximum of ca. 20% at 80 L/min. The power in both cases increases by a factor of 1.2: from around 5 kW at 30 L/min, to ca. 6 kW at 80 L/min. The energy cost for the process trends from a high value (around 11 MJ/mol at 30 L/min) to a low value as a function of flow rate, reaching a more viable value of 1.5 MJ/mol at 70 and 80 L/min.

For flow rates to the left of the peak (≤60 L/min), the post-plasma chamber is not sufficiently cooled by either the cold gas stream or the active wall cooling (or a combination of both). The maximum post-plasma temperatures recorded with these configurations coincided with a flow rate of 30 L/min, resulting in gas temperatures around 600 ˚C. While increasing the flow rate to 60 L/min did result in a temperature reduction to ca. 500 ˚C, this temperature was still too high for an optimum conversion performance. Taken together with the thermal data for three reactors (see above), the optimal temperature conditions correspond to the apparent measured gas temperature of around 450 oC (or lower, which could be achieved by more efficient cooling). This indicates that increasing the cooling capacity of the reactor after the plasma (hence further decreasing the post-plasma gas temperature) would reduce the extent of the recombination reactions between CO and O/O_2 to form the reactant CO_2. The energy cost of the process is significantly influenced by the conversion at each step. At low flow rates, especially 30 L/min, this low conversion results in a high energy cost. At high flow rates, such as 70 and 80 L/min, the dramatic increase in conversion leads to an improvement in energy cost. Thus, despite the low conversion at low flow rates (i.e., below 60 L/min), improvements to the energy cost can be obtained with a relative change in flow rate larger than a relative change in conversion. This is evident between 30 and 40 L/min, when the conversion increases by a factor of 1.2 while the flow rate increases by a factor of 1.3. At 70 and 80 L/min, the increasing conversion and flow rate work together to drastically decrease the energy cost of the process to a minimum value for these configurations.

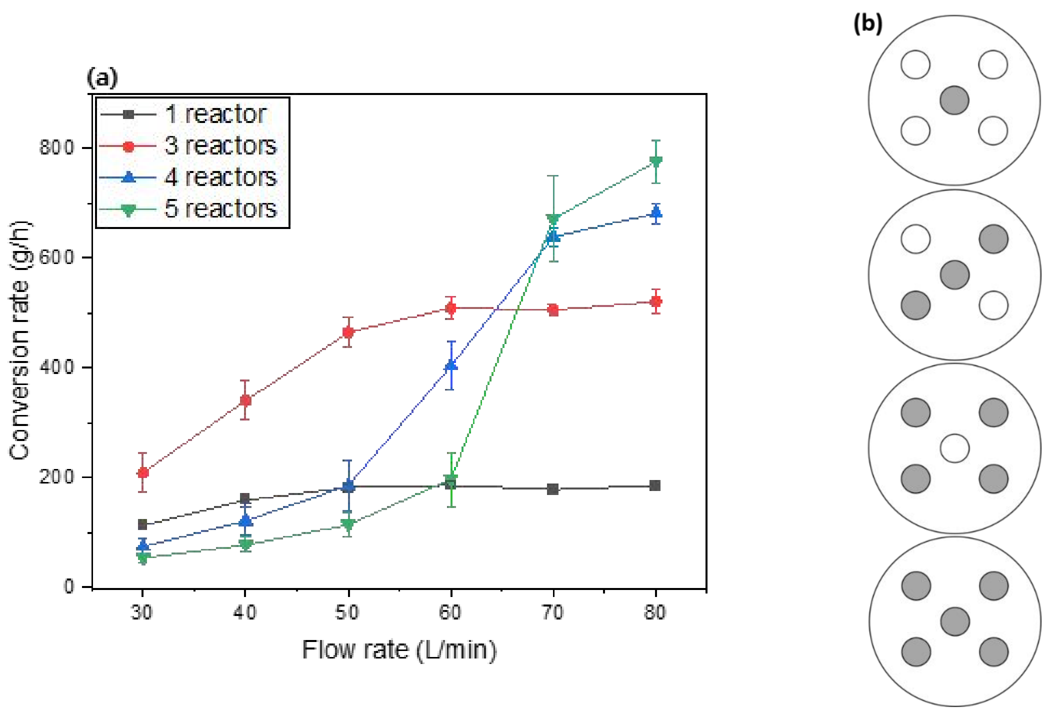

Figure S3. CO_2 conversion rate as a function of the feed gas flow rate **(a)** for various reactor configurations (**b**).

We compared the conversion rate (CR) between configurations with different numbers of active reactors (Fig. S3). When plotted as a function of the feed gas flow rate, the CR increases for all configurations investigated. For the single active reactor condition, while the absolute conversion decreases as a function of flow rate, it does not initially do so proportionally (between 30 and 50 L/min). This results in a slight increase in CR, reaching a steady value around 185 g/h for flow rates above 50 L/min. A similar trend is observed with three active reactors, although the CO_2 CR is larger, rising from ca. 200 g/h at 30 L/min to a steady maximal value around 500 g/h for flow rates ≥60 L/min. The trend of CR as a function of flow rate for four and five active reactors is quite similar, showing a steep rise between 50 and 70 L/min. For four active reactors, the CR increases by a factor of 3.7, while it increases by a factor of 6.8 for five active reactors. Importantly, the CR appears to plateau for three active reactors at around 60 L/min. Changing the flow rate from 70 to 80 L/min, with four reactors the relative increase in CR is lower than that with five reactors. This is another unambiguous indication that a larger number of active reactors yields best process metrics at higher flow rates. This is also clearly seen in Fig. 4 (in the main text), where the absolute conversion does not decrease when the flow rate increases from 70 to 80 L/min. In turn, this implies that increasing the flow rate even further could result in better process performance in the case of all five reactors.