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Citation

Smits, F. M. A., Bongers, W. A., Peeters, F. J. J., Deursen, C. F. A. M. van, & Sanden, M. C. M. van de. (2022). Efficiency and conversion optimisation of tubular vortex flow stabilised microwave plasma reactors. *48Th Eps Conference On Plasma Physics*, 560. Retrieved from <https://hdl.handle.net/1887/3514522>

Version: Publisher's Version

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Note: To cite this publication please use the final published version (if applicable).

Efficiency and conversion optimisation of tubular vortex flow stabilised microwave plasma reactors

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A total efficiency measure for a reactor with some energy efficiency and conversion rate is determined for a given CO separation enthalpy from a CO₂-CO-O₂ mixture. Including separation losses is shown to shift total efficiency from 40.3% at a flow rate which needs 3 times the input power for full conversion at 390 mbar, to 30.5% at a flow rate equivalent to 2 times the input power at 128 mbar. The efficiency and conversion losses are categorised in three components: slippage, heat loss and burn back. Slippage reduces the conversion rate, heat loss reduces the energy efficiency and burn back reduces both. The components are identified in the forward vortex microwave reactor configuration. The relative importance of the various components to the total reactor efficiency is discussed and schemes to improve total efficiency are proposed.

The forward vortex microwave plasma reactor is being studied as a promising candidate for conversion of CO₂ to CO as a pathway to convert electrical energy into chemical energy [1]. The output is a mixture so after the reactor, separation steps are required to remove oxygen and CO₂ from the desired product CO. When assessing microwave reactor efficiencies, the CO concentration at the output flow is measured and the chemical power in the output CO flow is compared to the input power. If we also include the energy required for the separation of CO from CO₂ and O₂, the system analysis results in the following relation for the total efficiency η_{tot} :

$$\eta_{tot} = \eta_{react} \frac{1}{1 + \frac{\Delta H_{sep}}{\Delta H_{conv}} \left(\frac{Q_{in} \Delta H_{conv}}{P_{in}} - \eta_{react} \right)}$$

Here ΔH_{sep} and ΔH_{conv} are the separation and conversion enthalpies per molecule, Q_{in} and P_{in} the input flow and power, and η_{react} is the reactor energy efficiency. When applying this relation to experimental efficiency data of the forward vortex reactor [2], the optimum operational conditions change considerably (Fig. 1). Solid lines are efficiencies excluding separation losses, dotted lines include separation losses. The upper number in the legend indicates

the flow rate, the lower number compares the power required to fully convert this flow to the input power. The enthalpy to separate CO from the output mixture is taken only 15% of the conversion enthalpy, corresponding to for example a pressure swing adsorption separator. Visibly, at increasing flow to power ratios, the total efficiency is increasingly reduced since larger amounts of CO₂ have to be removed from the CO. Without separation losses, the optimum efficiency is 40.3 % at 390 mbar for a flow with a conversion power of 3 times the input power. With separation losses, the optimum efficiency is 30.5 % at 128 mbar and a flow with conversion power of only 1.95 times the input power.

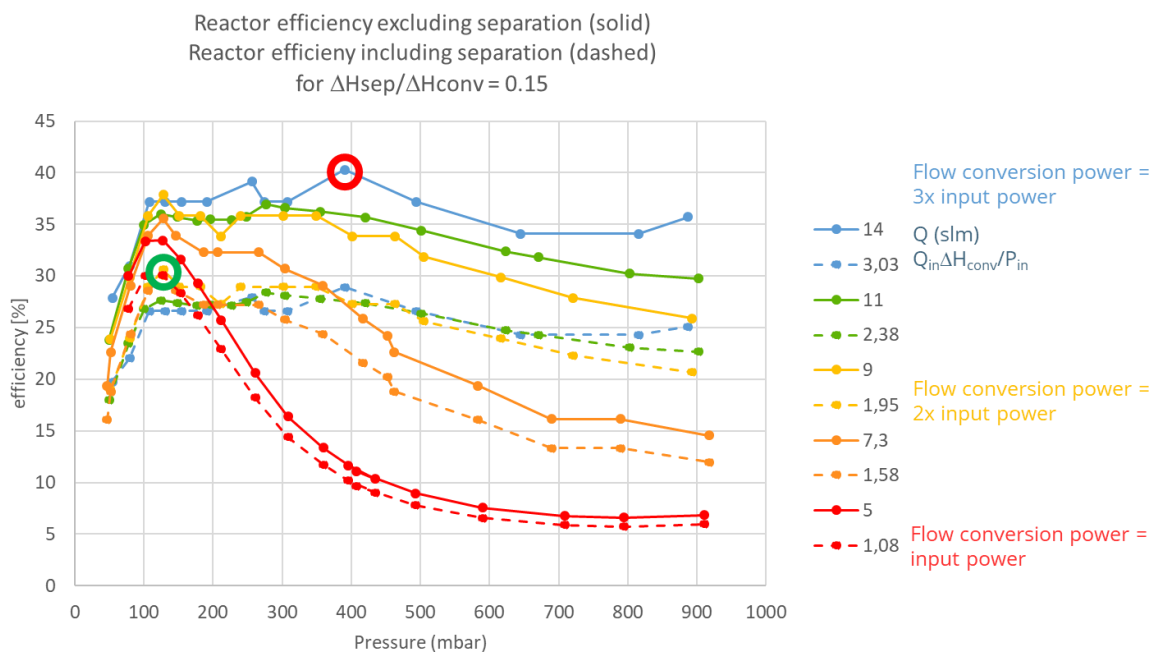


Fig. 1. Experimental energy efficiencies excluding (solid lines) and including (dotted lines) separation energy loss. Circles indicate the optimum conditions for the two cases.

To analyse the relations between efficiency and conversion, the mass flows and energy flows are summarized per reactor process in a single diagram and identified in a reactor simulation (Fig.2). In the diagram, mass flow is shown in vertical direction and energy flow in horizontal direction. The desired processes are heating until full dissociation in the hot zone, followed by molecular oxygen formation and fast cooling to freeze the reaction products. Freeze-in is established through mixing and diffusion with the surrounding gas. Many processes cause power losses and reduce total energy efficiency: microwave power generation and leakage, radiation, convection, tube conduction and cooling, output product heat, and separation. Also, besides the process flow region, a large slippage region is located in which gas is not processed and which reduces the conversion rate. Finally, some carbon monoxide is burning back to CO₂ while losing heat and decreasing the amount of converted CO₂, reducing both efficiency and

conversion rate. The simulations in the graph besides the diagram were performed assuming cylindrical symmetry, an elongated Gaussian central power density profile corresponding to optical emission measurements, and included flow, heating, heat transport, diffusion, chemistry, mixing and quenching. The simulation figure is widened by a factor of 2 for clarity. Flow paths in the cross-sectional plane are indicated in white. A large recirculation cell develops above the plasma of which the content does not contribute to the output. The flow paths besides the recirculation cell follow a heating trajectory through the heating region up to the dissociation region while part is not sufficiently heated and enters the slippage and mixing regions. After the dissociation region, the burn back and oxygen formation region is entered. Diffusion and flow from these regions into the surroundings cools the gas and fixes the output products. Along the tube, the mixing region grows until it fully covers the slippage region.

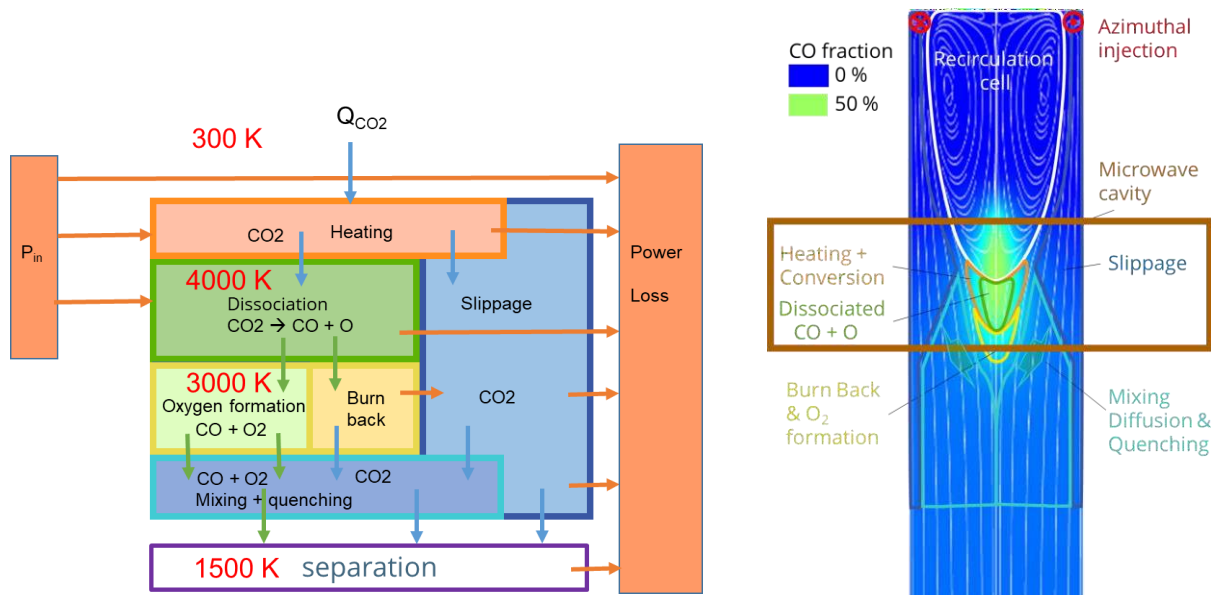


Fig. 2. Mass flow (vertical) and energy flow (horizontal) diagram (left) and reactor simulation (right) with flow lines (white) and CO fraction background colour (blue to green). Process regions are indicated in corresponding colours in the two diagrams.

To assess the impact on efficiency and conversion of the processes, we plot the input power versus flow (Fig.3) and multiply flow with conversion enthalpy so it represents the power needed to convert the flow to CO. If the input power equals the power needed to convert the inflow, the operational setting (A) is on the diagonal. If slippage occurs, this reduces the conversion. If heat loss occurs, this reduces the efficiency. Finally, burn back reduces conversion as well as efficiency with the corresponding heat losses. In case the reactor is operated at powers much higher than needed for conversion of the input flow (B), it imposes heat loss since the power cannot be spent on conversion, leading to low energy efficiency.

However, due to the large amount of energy, little flow will avoid the hot zone and conversion can be high. In case the reactor is operated at flows higher than the available power to convert the flow (C), the flow cannot be converted and additional slippage is imposed. However, due to the large amount of available CO₂, little heat will be lost and efficiency can be high.

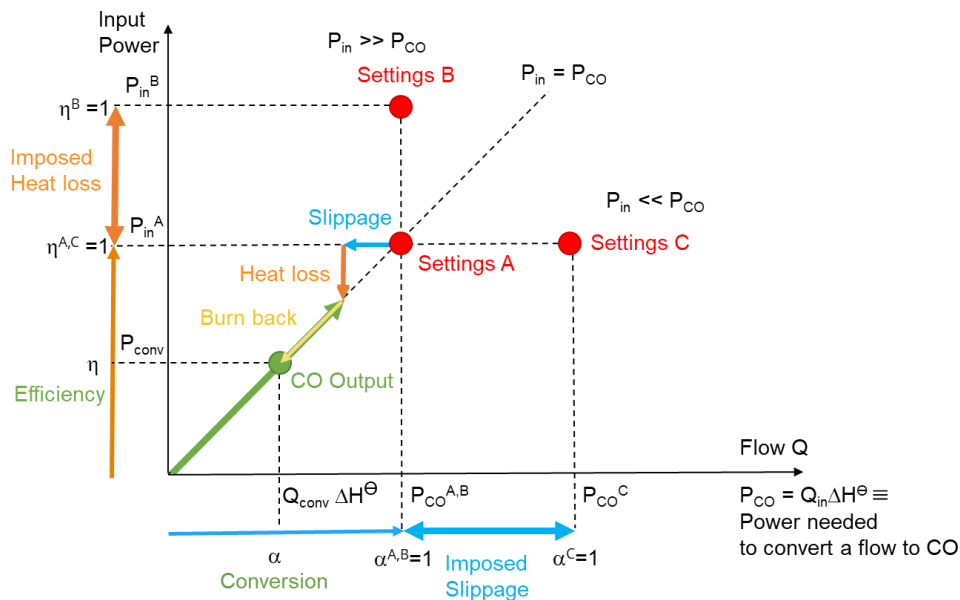


Fig. 3 Input power versus power to convert the inflow and the consequences for three different reactor operation settings

This basic analysis points to four directions to optimise both efficiency and conversion: Operate the reactor at powers are equal to the power required to convert the inflow to avoid heat loss and slippage imposed by the operational setting. Minimize burn back by designing fast product freeze in trajectories: this reduces both efficiency and conversion loss. Minimize slippage by a detailed design of the flow trajectories that forces all flow to be converted. Minimize heat loss by insulating the reactor and applying heat recovery to the hot output flow. Efficiency and conversion can also be optimised by promoting associative dissociation of the atomic oxygen with CO₂ which recovers the chemical energy of the atomic oxygen [3].

[1] Bongers W.A. et al., Plasma-driven dissociation of CO₂ for fuel synthesis, Plasma Processes Polym. 14 (2017) 1600126 - <https://doi.org/10.1002/ppap.201600126>

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[3] A.W. van de Steeg et al (2021) ACS Energy Letters 6, 2876, 2021