Improvement of Plasma Reactor Design for Methane Pyrolysis by Simulation

Aliya Magazova* and Prof. Dr. David W. Agar

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Hydrogen generation by plasma reactors from methane is a promising technology. Although, much research is done on accurately describing the plasma, fewer was focused on the reactor design optimization. By altering the design, the velocity profiles and heat transport can be managed in such a way that the conversion of methane can be increased. Through a simplified simulation approach, it was possible to increase the gliding arc reactor conversion from 30 to 50 %, which was tested by experiments. Similar finding resulted from the microwave plasma simulation, where simulations showed a potential rise from 66 % to 84 %.

Keywords: CFD simulation, green hydrogen, Methane pyrolysis, Plasma, Reactor design

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1 Introduction

One way to reduce the CO_2 emissions is by the use of sustainable energy sources. Hydrogen is a potential zero-emission fuel since no greenhouse gases are produced from hydrogen combustion. In order for hydrogen to be a feasible solution against the climate crisis, it should be generated through emission-free methodologies accordingly [1]. For instance, methane pyrolysis in an atmospheric plasma reactor powered by CO_2-free electricity source could be an attractive option from energy point of view, since the reaction enthalpy to split the methane molecule is several times lower than to split water molecule (Eq. (1) and (2)).

$$CH_4 \rightarrow 2H_2 + C\,\mathrm{kJ\ mol^{-1}} \tag{1}$$

$$H_2O \rightarrow H_2 + 0.5O_2\,\mathrm{kJ\ mol^{-1}} \tag{2}$$

Plasma involves the conversion of neutral species into positive ions and electrons and radicals. Through electrons collision with molecules, the energy is transferred and propagates ionization, excitation, dissociation, and chemical reactions. The attributes of plasma reactors, which comprise of elevated temperatures of up to 10 000 K, high concentrations of active species, electric and magnetic fields that can improve the mass transport, make this technology appealing. Two examples of such reactors are gliding arc plasma (GAP) and microwave plasma (MW) reactors. The key features of the GAP reactor are that the arc is ignited between electrodes by electric field, high temperatures at the arc and gas is at 300–1000 K, and the substantial electron density $n_e \geq 10^{20}\,\mathrm{m^{-3}}$ [2]. The microwave plasma, on the other hand, treats larger gas volumes with the gas temperature at 4300 K, and plasma is produced through magnetron with 2.45 GHz frequency [3].

However, plasma and its properties are extremely complex with the research still on-going. Moreover, the accurate simulation is a time-consuming task, due to number of species involved in the simulation and the range between the reaction time (µs) and residence time (s). As a consequence, an identification and focus on the few main parameters, which influence the plasma pyrolysis and methane conversion, is necessary to model and simulate a plasma reactor. Specifically, simulations open the potential for improvement of reactor design. Thus, a systematic approach including the replacement of plasma as a heat source is used, so that the reactor could be optimized in a limited amount of time [4]. 3D simulation of the GAP reactor reveals that the vortex length, velocity and intensity can positively influence the conversion of methane. And the simulation of MW reactor shows that methane feed location can increase methane conversion. There are few studies regarding the plasma reactor geometry optimization [5–7], whereas most of it is dedicated to the exactness of the plasma description. In this paper, we present the insights gained by the simulation for reactor optimization and the application in laboratory experiments.

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[1] Aliya Magazova (aliya.magazova@tu-dortmund.de),
[1] Prof. Dr. David W. Agar

[1] Technische Universität Dortmund, Lehrstuhl für Chemische Verfahrenstechnik, Emil-Figge-Straße 66, 44227 Dortmund, Germany.

2 GAP Reactor Design Improvements Based on Simulations

The original geometry of GAP reactor was based on literature [8]. The outlet tube is an anode, the cap is a cathode, and the gas enters through the tangential inlets. Initially, the gas would swirl in the vortex generation ring before entering the cathode cap, as shown in Fig. 1a. The arc is then created in the smallest distance between the electrodes and is carried to the reactor center by the flow. After closely observing the velocity vectors in 2D, it is distinct that the flow is obscured by eddies, which, in fact, may hinder the arc integrity. In comparison, Fig. 1b demonstrates the positive influence of removal of the vortex generation ring on the flow pattern. It is noticeable that the gas enters the reactor following a straight line and can push the arc towards the center. The eddies in Fig. 1a can result in short-circuit, if the gas speed is dissipated due to eddies and the arc stays unmoved for a longer period of time in a confined space.

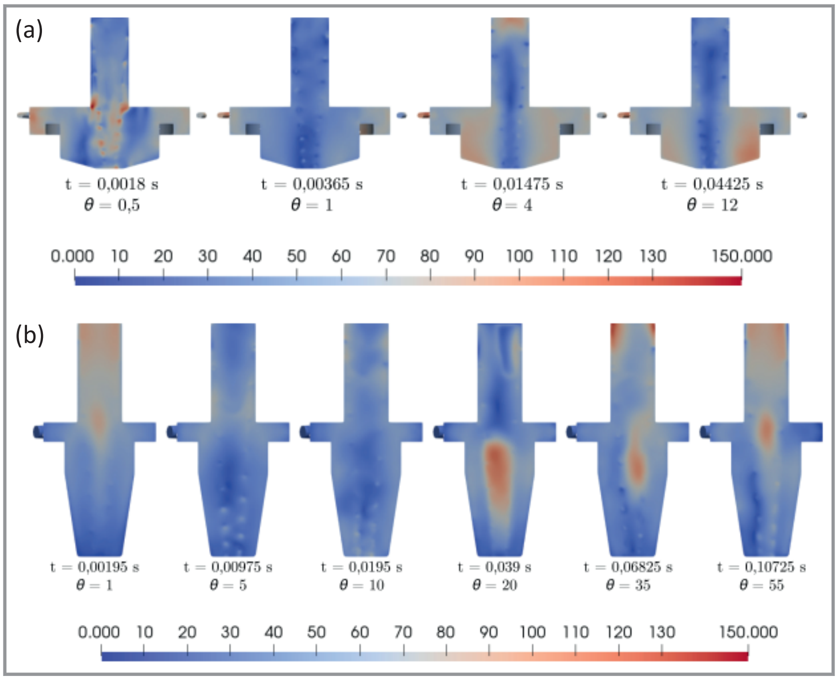


Figure 2. The development of the velocity profile (m s^{-1}) of the original (a) and alternative (b) GAP reactor including the impact from the heat source and chemical reactions.

The simulated design alteration was tested in [3], 31–35 % conversion was achieved, which is comparative to the results from the original source [9]. It means that the outcome is not jeopardized. Such elimination of redundant parts also reduces the computational time, because there are less mesh cells to solve and the convergence is faster due to lower turbulence of the flow.

Another observation from the experiments is that if the vortex is stable and expands evenly along the reactor, the plasma arc can be concentrated more in the middle of the reactor and maintained easier. Additionally, the heat losses are reduced due to more efficient energy distribution. Therefore, the new geometry possesses a longer reactor body that also results in lengthened plasma arc. The body of the reactor is narrower, which helps to stabilize the vortex. Fig. 2b demonstrates that for the same volumetric gas flow, the velocity in case of a longer geometry is lower. That

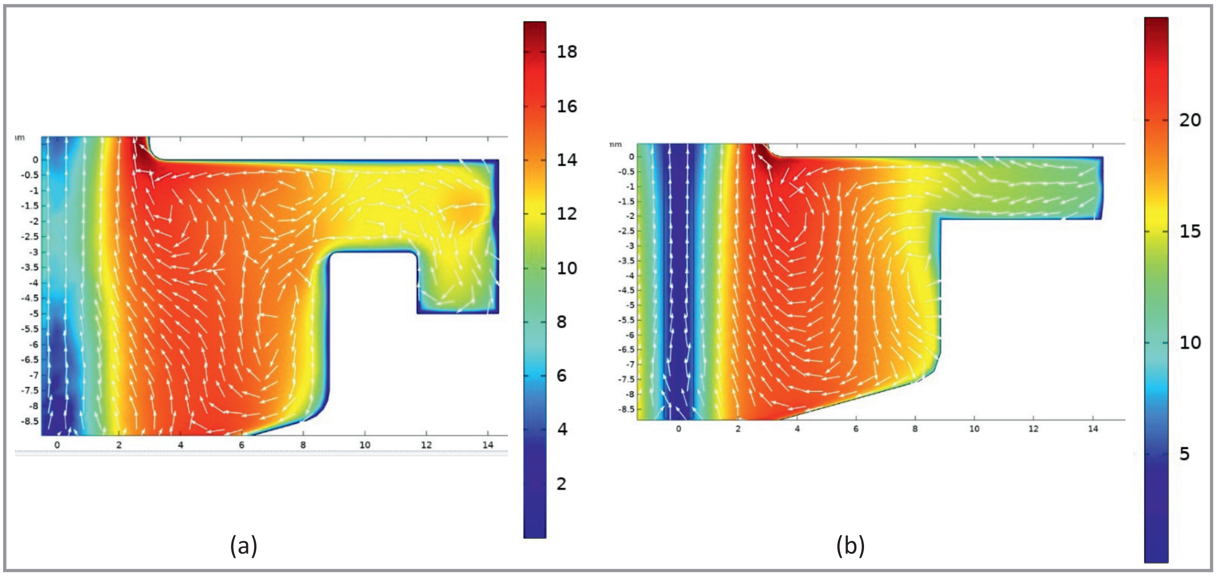


Figure 1. 2D cut of the velocity field (m s^{-1}) of the original GAP reactor (a) and modified GAP reactor (b).

implies that the residence time for the alternative geometry is longer. Hence, gas conversion can increase.

To note, the length of the reactor is proportional to the arc size, which, indeed, determines the power consumption. In other words, the larger arc needs more power input. Therefore, the length of the reactor is limited to the power supply available.

In Fig. 3 one can see that narrower GAP reactor leads to more efficient heat distribution. The reactive zone region is amplified, which can enhance the methane conversion. The experiments showed that the altered GAP geometry raised the methane conversion from 30 % to 50 % [10].

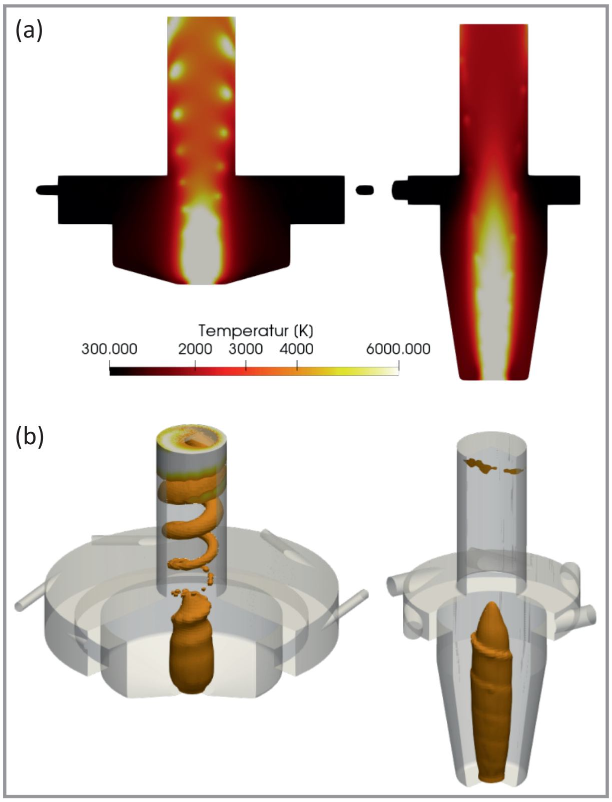


Figure 3. Comparison of steady-state temperature profiles of the original (left) and new geometry (right) of the GAP reactor (a). Temperature greater than 4000 K is highlighted in (b).

3 MW Reactor Design Improvements Based on Simulations

MW reactor design consists of an outlet quartz tube and tangential inlets at one end of the tube. The magnetron waveguide is directed normally to the quartz tube, where plasma is ignited, and then carried further by the gas. Fig. 4 shows the transition between MW plasma in the lab to the simulation. Fig. 4a is a plasma photo through the observation window. The lightest portion of the photo indicate the highest temperature in the tube. For the simplification, plasma was substituted by a heat source with a bell-like normal distribution shape (Fig. 4b), so that the reactor design could be optimized. The simulated temperature profile is also given in Fig. 4, and the location of plasma is clearly demonstrated.

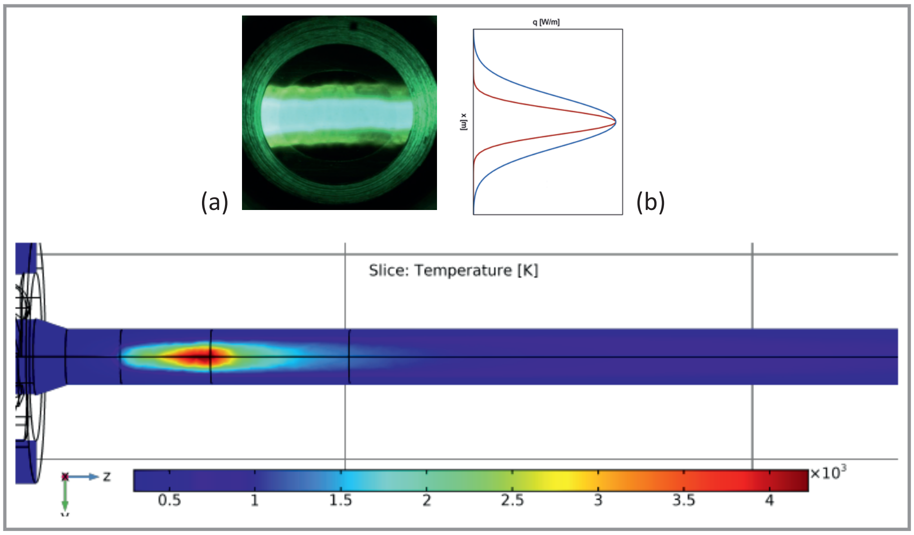


Figure 4. Microwave plasma (a) temperature profile resembles normal distribution function (b). The maximum temperature is in the center of the reactor discharge tube, and it coincides with magnetron location (below).

The simulation of the temperature profile was adjusted by experimental results to compensate for the assumption of the plasma as a heat source, so the simulated reactor optimizations could be applied to the experiments. For this purpose, the normal distribution function was adjusted respectively. Fig. 5 shows that the simulated and actual temperature profiles are similar. Hence, the next step of reactor design optimization could be carried out. Similarly, the reaction mechanism was set in a way to include the important reactions so that the computational time is small, however, the experimental results are still resembled. Fig. 6 show that a reasonable reaction set is selected with minor deviation at some operational conditions. Fig. 5 shows that with increasing the power supply, the temperature increases as well. Also, with higher CH_4 ratio, due to its reactivity in comparison to Ar, temperature rises.

According to Fig. 4, most of the heat is located in the outlet tube center. Therefore, methane conversion could be increased if more of the effluent would pass through the center. Hence, the idea of feeding the reactive gas, methane, through the

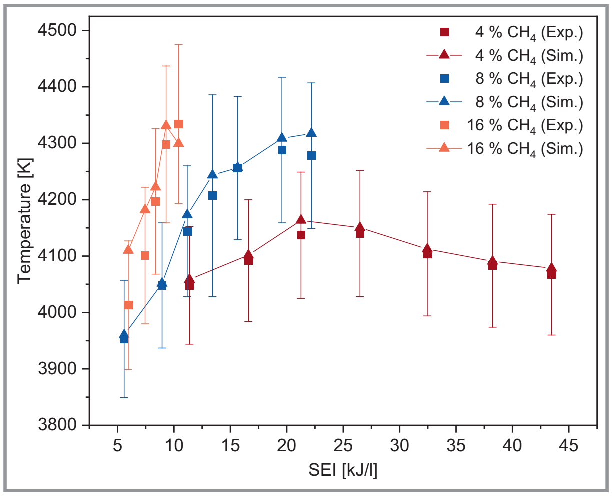


Figure 5. Maximum temperature in the experiments compared to the simulations (at 70 slm).

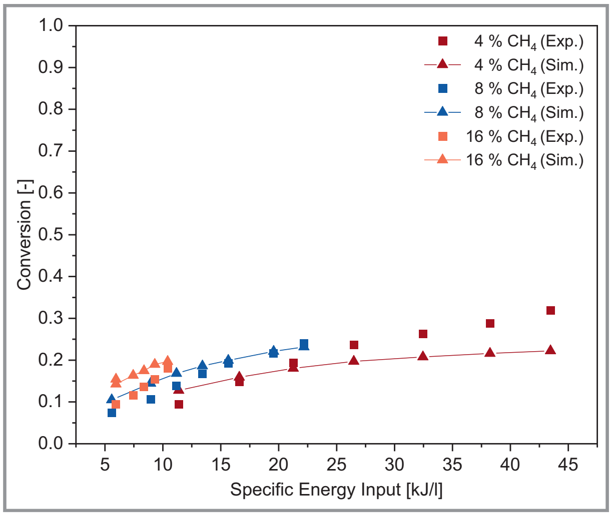


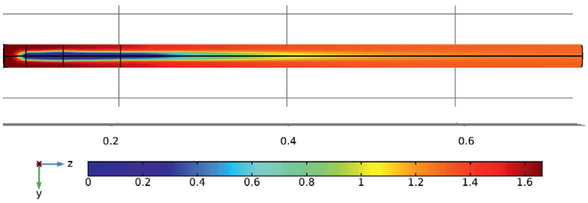
Figure 6. Methane conversion in the experiments compared to the simulations (at 70 slm).

center, and the carrier gas, argon, through the tangential inlets was simulated.

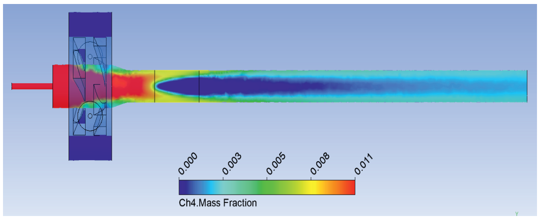
Fig. 7a shows the concentration of methane in the outlet tube, where all gas is fed through the tangential inlets. It is evident from the high methane concentrations near the tube walls that that large amount of gas passes around the hot zone without reaction. In Fig. 7b, where methane enters through the center, the chance of methane going through a zone of high temperature is higher. Thus, the simulated conversion of methane is increased from 66 % to 84 %.

4 Conclusion

The use of simulations and adequate simulation approach showed that it is possible to spot the possible reactor design improvements. The suggested approach involves the adaptation of the plasma as a heat sources, as well as the reduction of reaction set. Nevertheless, the base simulation is checked against the experimental results, so that the suggested design improvements are reliable. Simulations open the weaknesses of the original design from the velocity profiles and temperature distributions. Therefore, this shows that even with simplified simulation approach, without heavy details, working design optimization is possible.



(a) X_{CH4} = 66 %



(b) X_{CH4} = 84 %

Figure 7. Comparison of CH_4 concentrations (mol m^{-3}) CH_4 inlet modification for microwave plasma reactor: a) CH_4 together with argon carrier gas enters the reactor through tangential inlets, b) CH_4 is fed through the central inlet and argon carrier gas through tangential ones.

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Symbols used

ΔH	[kJ mol^{-1}]	heat of reaction
n_e	[m^{-3}]	electron density
X_{CH4}	[%]	methane conversion

Abbreviations

GAP	gliding arc plasma
MW	microwave

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