https://doi.org/10.46813/2021-131-131

SIMULATION OF GAS DYNAMICS IN PLASMA REACTOR FOR CARBON DIOXIDE CONVERSION

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Numerical simulation of a bulk-type plasma reactor for carbon dioxide conversion with distributed gas injection and pumping has been performed in hydrodynamic approximation by solution of Navier-Stokes equation using the mathematical package COMSOL. It is shown that the geometry of gas injection and pumping, which determines the trajectories of the particles and their residence time in reactor, can significantly affect the energy efficiency of the conversion. Different particles on their way from inlet to pumping hole move along different trajectories and spend different times inside the reactor. If the residence time of the gas in the reactor is less than optimal, the gas conversion will be incomplete. If this time is more than optimal, then an excessive amount of energy will be applied to the already converted gas. It is shown that the reactor height affects significantly the energy efficiency of plasma conversion of carbon dioxide.

PACS: 52.77.-j

INTRODUCTION

The increase in carbon dioxide emission due to anthropogenic impact leading to global warming is one of the most serious challenges for humanity in the 21st century. CO_2 accumulation can be avoided only through processes that quickly and efficiently bind carbon dioxide, preferably converting it into fuel or useful chemicals. One of the promising approaches in this field, which is not yet used on an industrial scale, is the plasma technology. Plasma conversion of carbon dioxide possesses some important advantages in comparison to classical thermal CO_2 splitting and is actively researched [1-10] with use of different kinds of gas-discharge plasma: glow discharge [1], dielectric barrier discharge (DBD) [2, 9, 10], helicone discharge [4], microwave discharge [8].

In [11, 12] the results of experimental study of conversion efficiency of CO_2 into CO and O_2 in various gas discharge devices at low gas pressure of 1...100 mTorr are presented. A comparative analysis of RF discharges of inductive and capacitive types, discharge in crossed EH fields with anode layer, magnetron discharge was performed. The general result of these studies is the fact that achievement of high conversion ratio of carbon dioxide of about 80 % is possible for many types of discharge, but the energy efficiency of conversion is always low (as a rule, it is only a few percent). The maximum achieved conversion factor was 86 %, while the typical values of the energy efficiency were 1...3 %; only in the case of RF inductive discharge of very low power it was up to 50 %.

One of the reasons for the low energy efficiency is the nature of the gas-discharge plasma, in which the energy of the external field deposited to electrons is expended through many channels, and only the energy to dissociation channel is useful. In [13], the results of calculations of the efficiency of energy use for CO_2 molecule dissociation in gas-discharge plasma are presented. From these results it can be concluded that in the region of high values of reduced electric field E/N (more than 200 Td) the main conversion channel is the electron impact dissociation. In this case, the share of energy that goes to dissociation does not exceed a few percent.

However, there is another important factor that can significantly affect the energy efficiency of the conversion. Different particles on their way from inlet to pumping hole move along different trajectories and spend different times inside the reactor. At the same time, for each value of input power and of carbon dioxide gas flow, there is an optimal time of interaction of gas with plasma. If the residence time of the gas in the reactor is less than optimal, the gas conversion will be incomplete. If this time is more than optimal, then an excessive amount of energy will be applied to the already converted gas.

The shape and volume of a plasma reactor are often determined by the gas discharge used. For high pressure reactors (e.g. atmospheric), these are often narrow slits or tubes. However, at lower gas pressures it is possible to use bulk type reactors, where the plasma occupies a fairly large space. For such reactors, one of the key issues is the geometry of the gas injection and pumping, which determines the trajectories of the particles and the time they stay in the reactor. The study presented in this article is devoted to research of this issue for a bulk-type reactor with distributed gas injection and pumping.

1. MODEL DESCRIPTION

This paper considers the simplest case of a cylindrical reactor with metal electrodes at the ends of the chamber and with dielectric side wall. As the reactor will use an discharge with electrodes (DC, RF, pulsed), then one of the electrodes must be high-voltage and the other is grounded. Due to the possibility of parasitic discharges, the gas feeding and pumping must be carried out from the side of the grounded electrode. Due to the need to perform the conversion uniformly through-

ISSN 1562-6016. ВАНТ. 2021. №1(131)

out the chamber, it is necessary that the gas feeding and pumping to be distributed on the surface of the electrode.

The geometry of the reactor is presented in Fig. 1. The chamber diameter was 55 mm. The calculations were performed for two different camera lengths: 30 and 10 mm (hereinafter these two cases will be referred to as "long" and "short" chambers). The inlet and outlet openings are located on a regular hexagonal grid with a step of 5 mm along the diagonals. The total number of inlet openings was 72, and there were 19 pumping holes (Fig. 2). The diameter of all the holes was 1 mm.

Carbon dioxide pressure in the chamber was chosen to be 5 Torr (close to this pressure is observed near the surface of Mars, whose atmosphere is 95 % carbon dioxide). Since the largest Knudsen number was 0.01 in the holes, we applied the hydrodynamic approximation and solved the Navier-Stokes equation using the mathematical package COMSOL version 5.5 with a physical interface "Laminar Flow". The simulation was performed for carbon dioxide mass flow of 10 sccm.

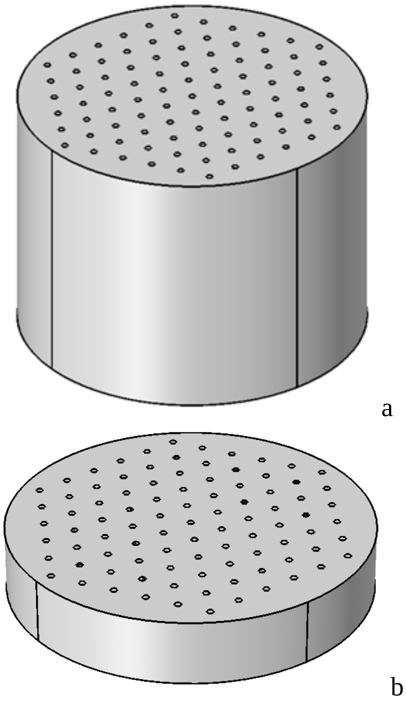

Fig. 1. General view of the simulatioj model of long (a) and short (b) chamber of the plasma-chemical reactor

The problem was solved in three-dimensional geometry with a partition of the solution area using a tetrahedral mesh with an adaptive step. Near the holes, the grid division step was about 0.1 mm, gradually increasing to 2 mm in the chamber volume. The total number of subdivisions was about 2.5 million.

The result of the numerical solution was the spatial distributions of pressure and gas velocity. Using the velocity field, the trajectories of the particles were reconstructed, with further analysis of distribution of particles by the time spent in the chamber and by trajectory lengths. The number of trajectories along which the distributions were constructed was at least 50 thousand that ensured comparative smoothness of the distributions. The initial coordinates of the trajectories were chosen randomly with the control of the radial density of points. Gas velocity profile in the inlet channel is defined by Poiseuille's law, thus, the distribution of the starting points of the trajectories was chosen in such a way that the point density was proportional to the gas velocity at a given radius.

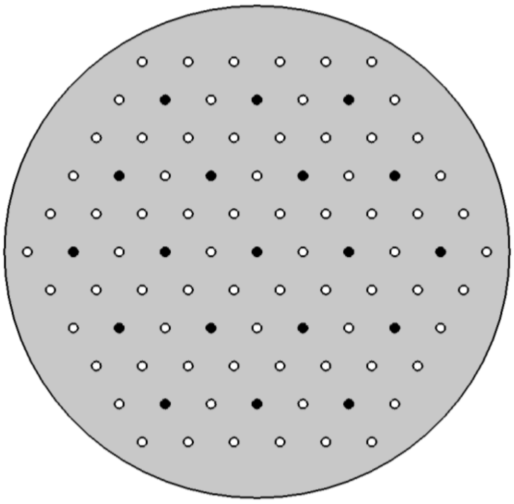

Fig. 2. Layout of inlet and outlet holes. Inlet holes are marked in white, pumping holes are in black

2, SIMULATION RESULTS

Fig. 3,a shows the variety of particle trajectories in the long chamber. For better clarity Fig. 3,b,c show a top view and a cross section of the chamber passing through its axis. From the top view it becomes clear that the trajectories of particles are significantly curvilinear, and they don't lie in a vertical plane. One can see different types of the trajectories: some of them connect adjacent holes while another travels for longer path.

In Fig. 3,c three groups of particles are well visible, namely: the particles passing in the nearest pumping hole; those pumped by the second row of holes; and those that go to the central pumping hole after a long stay in the bulk of the chamber.

It can be assumed that to reduce the useless energy losses, it is necessary to reduce the chamber length, limiting the possibility of the existence of trajectories of particles of the second and third groups. To test this hypothesis, similar calculations were carried out for the short chamber (10 mm long). The simulation results are shown in Fig. 4. It can be seen that in the short chamber the trajectories of the particles are more often closed to the adjacent exhaust holes, and the particles of the second and third groups are much less numerous than in the long chamber. This circumstance allows us to assert that the particles leave the volume faster and in a more grouped manner, which should have a positive effect on the energy efficiency of conversion.

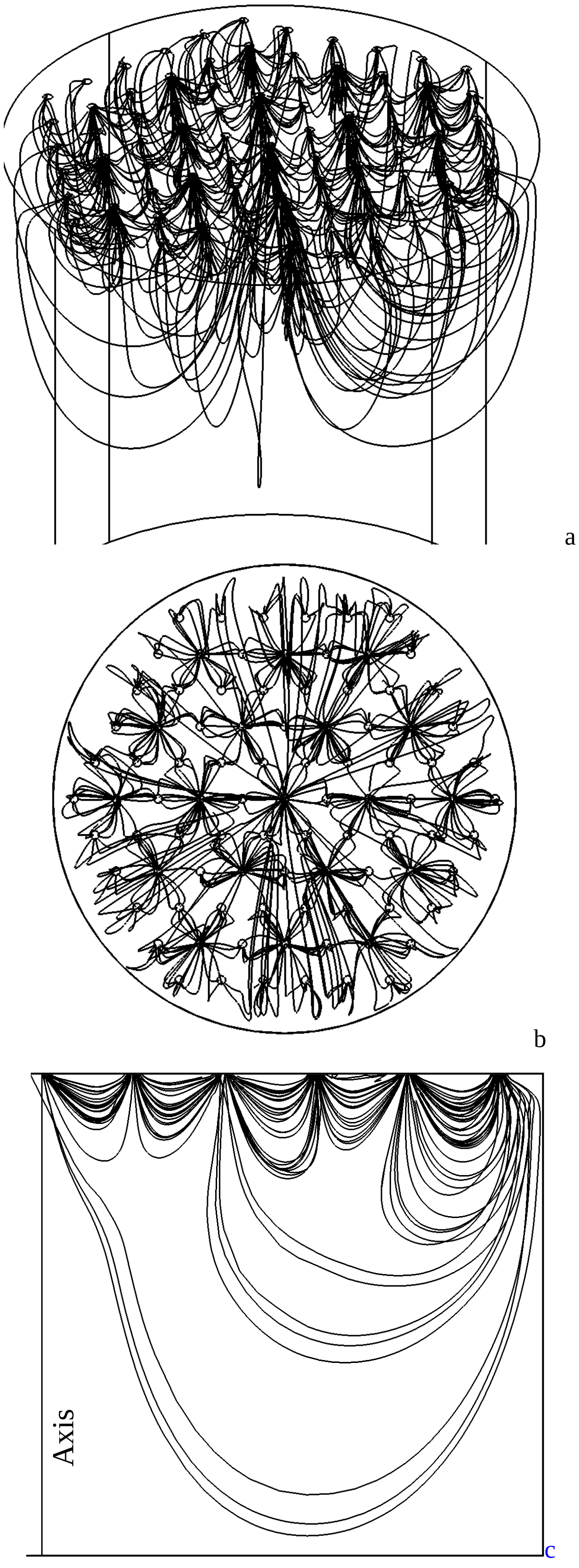

Fig. 3. Trajectories of particles in the long chamber: a – general view; b – top view; c – cross-section

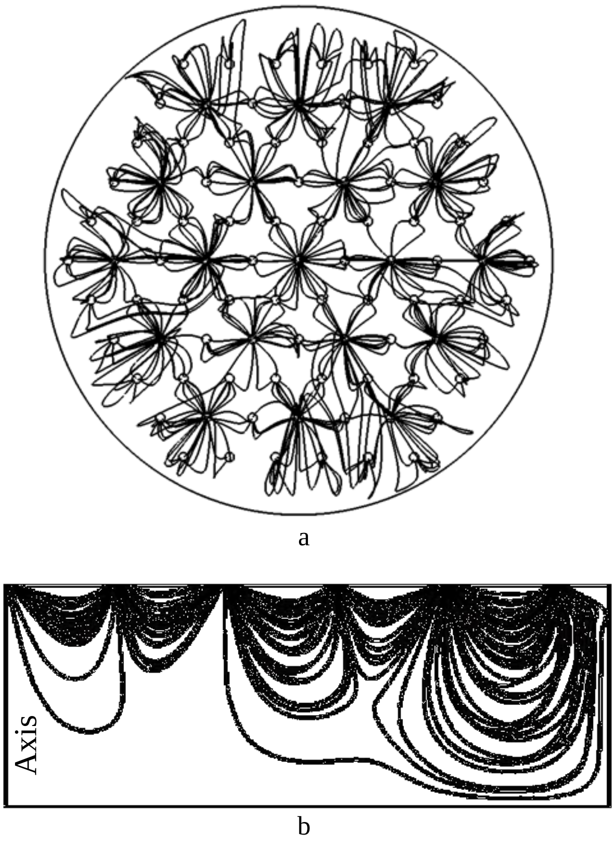

Fig. 4. Trajectories of particles in short chamber: a – top view; b – cross-section

Figs. 3, 4 show that the trajectories of the particles are very diverse and substantially curvilinear, that requires the use of a statistical study of the characteristics of particle motion.

In Fig. 5 shows the distribution functions of particles by trajectory length for the long and short chambers. It can be seen that the distributions for the first group of particles are close for both chambers, while in the long chamber there are much more particles with long trajectories, which confirms the assumption made.

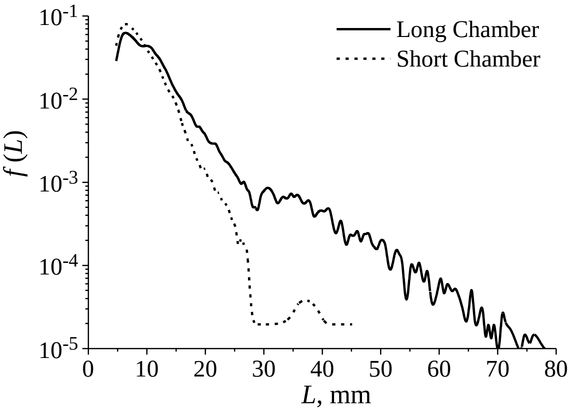

Fig. 5. Distribution function of particles by trajectory length

To find the energy absorbed by a particle, the residence time of the particle in the reactor is important rather than the trajectory length. Analysis of the calculated spatial distribution of the velocities shows that the particle velocity can vary by orders of magnitude along the trajectory. The velocity is maximum near the inlet and outlet holes, while in the lower part of the chamber the velocity is close to zero. Therefore, the velocities of all particles were integrated along the trajectories, and the distribution function of the particles by the residence time in the reactor was built, which is shown in Fig. 6. The figure confirms the difference between the chambers stated above, which consists in the greater portion of long-lived particles in the long chamber.

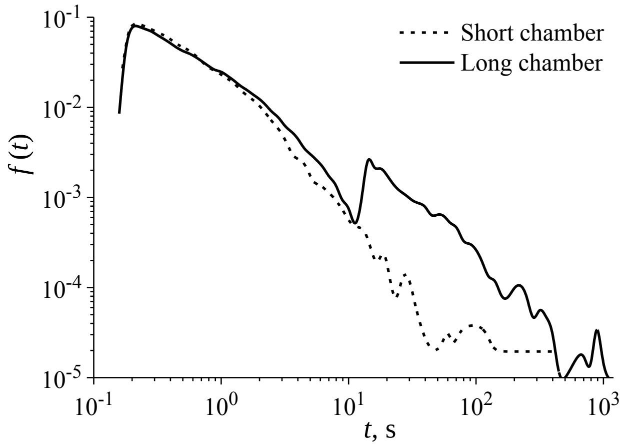

Fig. 6. Distribution function of particles by residence time in the chamber

One feature of the distribution in Fig. 6 attracts attention. While the lengths of the trajectories are distributed in a not very wide interval of 5...80 mm, the time distribution is 4 orders of magnitude wide. The travel time of particles of the second and third groups from the entrance to the exit can be as long as hours. During such a long time, the particles will absorb a huge amount of energy, and these expenses are useless. Since the absorbed energy is proportional to time, in order to estimate the contribution of various groups of particles to the energy efficiency of the conversion, let us introduce function $\mathcal{E}(t) = t\cdot f(t)$, which is a measure of energy absorbed by a particle. The calculated function $\mathcal{E}(t) = t\cdot f(t)$ is plotted in Fig. 7.

It is obvious from the figure that, despite the fact that the third group of particles in the long chamber is small compared to the first group, the energy deposited to the third group is greater than the energy input to the particles of the first group. Particles of the second and third groups, on average, are converted in the same time as the particles of the first group, but being in the volume of the chamber much longer than the conversion time. Conversion products absorb the input power, taking part in such processes as excitation, heating, ionization and others. Thus, the energy efficiency of carbon dioxide conversion decreases dramatically.

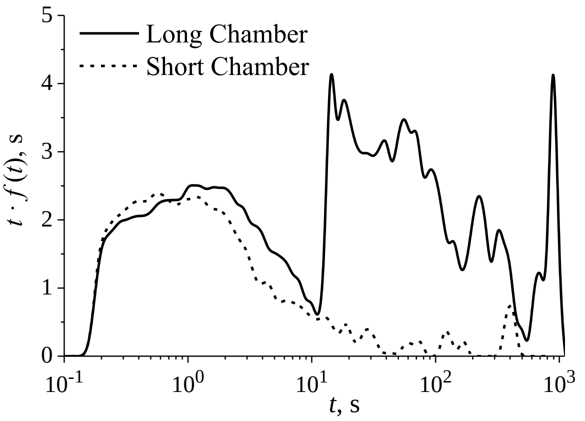

Fig. 1. Calculated distribution function $\mathcal{E}(t) = t\cdot f(t)$, which is a measure of energy absorbed by a particle

CONCLUSIONS

In the present paper, numerical simulation of a bulk-type plasma reactor for carbon dioxide conversion with distributed gas injection and pumping has been performed in hydrodynamic approximation by solution of Navier-Stokes equation using the mathematical package COMSOL.

It is shown that in the reactor with distributed gas injection and pumping different groups of particle trajectories exist with different particle residence times. Different particles on their way from inlet to pumping holes move along different trajectories and spend different times inside the reactor. If the residence time of the gas in the reactor is less than optimal, the gas conversion will be incomplete. If this time is more than optimal, then an excessive amount of energy will be applied to the already converted gas. We have shown that the reactor height affects significantly the energy efficiency of plasma conversion of carbon dioxide. Thus, the geometry of gas injection and pumping, which determines the trajectories of the particles and the time they stay in the reactor, determines the energy efficiency of the conversion. To obtain greater efficiency of the plasma-chemical process one need to minimize the height of the plasma-chemical reactor. Particles of the first group leave the plasma in a short time, which is not much variable, and in order to increase the conversion coefficient, gas flow parameters can be selected in such a way that during the residence time of the particle in the reactor it will be converted receiving not so much excessive energy. In order to prevent the decrease in the energy efficiency of the conversion, it is necessary to avoid the appearance of long-living particles in the reactor, that can be achieved only if the length of the reactor does not significantly exceed the hole pitch.

REFERENCES

1. D. Wu, R.A. Outlaw, and R.L. Ash. Extraction of oxygen from CO_2 using glow-discharge and permeation techniques // *J. Vac. Sci. Technol. A*. 1996, v.14, № 2, p. 408-414.

2. L.M. Zhou, B. Xue, U. Kogelschatz, and B. Eliasson. Nonequilibrium Plasma Reforming of Greenhouse Gases to Synthesis Gas // *Energy and Fuels*. 1998, v. 12, p. 1191-1199.
3. B. Hu, C. Guild, S.L. Suib. Thermal, electrochemical, and photochemical conversion of CO_2 to fuels and value-added products // *Journal of CO_2 Utilization*. 2013, v. 1, p. 18-27.
4. Ch. Berenguer and K. Katsonis. Global Modeling of CO_2 Discharges with Aerospace Applications // *Advances in Aerospace Engineering*. 2014, Article ID 847097, 17 p.
5. T. Kozak and A. Bogaerts. Splitting of CO_2 by vibrational excitation in non-equilibrium plasmas: a reaction kinetics model // *Plasma Sources Sci. Technol.* 2014, v. 23, № 4, p. 045004.
6. Ch. De Bie, J. van Dijk, A. Bogaerts. The Dominant Pathways for the Conversion of Methane into Oxygenates and Syngas // *J. Phys. Chem. C*. 2015, v. 119, p. 22331-22350.
7. A. Bogaerts, T. Kozak, K. van Laer, and R. Snoeckx. Plasma-based conversion of CO_2: current status and future challenges // *Faraday Discuss.* 2015, v. 183, p. 217-232.
8. G. J. van Rooij et al. Taming microwave plasma to beat thermodynamics in CO_2 dissociation // *Faraday Discuss*. 2015, 16 p., DOI: 10.1039/c5fd00045a.
9. R. Aerts, W. Somers, and A. Bogaerts. Carbon Dioxide Splitting in a Dielectric Barrier Discharge Plasma // *Chem. Sus. Chem*. 2015, v. 8, p. 702-716.
10. R. Snoeckx, S. Heijkers, K. Van Wesenbeeck, S. Lenaerts, and A. Bogaerts. CO_2 conversion in a dielectric barrier discharge plasma // *Energy Environ. Sci*. 2016, v. 9, p. 999-1011.
11. S.V. Dudin, A.N. Dakhov. Conversion of carbon dioxide in low-pressure plasma // *Problems of Atomic Science and Technology. Series “Plasma Physics”.* 2018, № 6, p. 194-197.
12. S.V. Dudin, A.V. Zykov, S.D. Yakovin. Plasma assisted conversion of carbon dioxide in low-pressure gas discharges // *Problems of Atomic Science and Technology*. 2019, № 4, p. 141-146.
13. R. Snoeckx, A. Bogaerts. Plasma technology – a novel solution for CO_2 conversion? // *Chem. Soc. Rev*. 2017, v. 46, p. 5805-5863.

Article received 15.01.2021

МОДЕЛИРОВАНИЕ ГАЗОВОЙ ДИНАМИКИ В ПЛАЗМЕННОМ РЕАКТОРЕ ДЛЯ КОНВЕРСИИ УГЛЕКИСЛОГО ГАЗА

П.П. Платонов, С.В. Дудин, В.А. Лисовский

Численное моделирование плазменного реактора объемного типа для конверсии диоксида углерода с распределенными напуском и откачкой газа выполнено в гидродинамическом приближении путем решения уравнения Навье-Стокса с помощью математического пакета COMSOL. Показано, что геометрия напуска и откачки газа, определяющая траектории частиц и время их пребывания в реакторе, может существенно влиять на энергетическую эффективность конверсии. Различные частицы на своем пути от входа к выходу движутся по разным траекториям и проводят разное время внутри реактора. Если время пребывания газа в реакторе меньше оптимального, конверсия газа будет неполной. Если это время больше оптимального, то в уже преобразованный газ будет вложено чрезмерное количество энергии. Показано, что высота реактора существенно влияет на энергоэффективность плазменного преобразования углекислого газа.

МОДЕЛЮВАННЯ ГАЗОВОЇ ДИНАМІКИ В ПЛАЗМОВОМУ РЕАКТОРІ ДЛЯ КОНВЕРСІЇ ВУГЛЕКИСЛОГО ГАЗУ

П.П. Платонов, С.В. Дудін, В.О. Лісовський

Чисельне моделювання плазмового реактора об'ємного типу для конверсії діоксиду вуглецю з розподіленими напуском і відкачуванням газу виконано в гідродинамічному наближенні шляхом вирішення рівняння Нав'є-Стокса за допомогою математичного пакета COMSOL. Показано, що геометрія напуску і відкачування газу, що визначає траєкторії частинок і час їх перебування в реакторі, може істотно впливати на енергетичну ефективність конверсії. Різні частинки на своєму шляху від входу до виходу рухаються за різними траєкторіями і проводять різні часи всередині реактора. Якщо час перебування газу в реакторі менший оптимального, конверсія газу буде неповною. Якщо цей час більший оптимального, то в уже перетворений газ буде вкладена надмірна кількість енергії. Показано, що висота реактора істотно впливає на енергоефективність плазмового перетворення вуглекислого газу.