Atmospheric pressure glow discharge: design improvement based on modelling and experiments

G Trenchev[1], A. Nikiforov[2], W. Wang[1], St Kolev[3] and A Bogaerts[1]

[1]Research group PLASMANT, Department of Chemistry, University of Antwerp, Universiteitsplein 1, B-2610 Antwerp, Belgium
[2]University of Gent, St. Pietersnieuwstraat 33, 9000 Gent
[3]Faculty of Physics, Sofia University, 5 James Bourchier Boulevard, 1164 Sofia, Bulgaria

Abstract: A novel atmospheric pressure glow discharge (APGD) reactor is investigated for the purpose of plasma-assisted CO_2 conversion. The reactor is gradually improved, based on computer simulations, leading to three different configurations. An enhancement of the conversion performance is indeed observed experimentally. The third design, so-called "Confined APGD" delivers a CO_2 conversion of up to 12.5%, at energy efficiency around 25%. The plasma processes are investigated by means of plasma fluid dynamics modelling.

Keywords: plasma, modelling, CO_2, atmospheric pressure glow discharge, dc discharge, vortex, conversion, energy efficiency, reactor design

1. Introduction

Atmospheric pressure glow discharges (APGDs) are DC non-thermal plasma sources existing in a variety of different configurations. These devices are characterized by their robustness and ease of application, especially in the field of plasma-assisted CO_2 conversion, due to their ability to work at atmospheric pressure. They produce a low-temperature glow-like plasma with moderate density and relatively high electron temperature. The APGD is a low-current, steady DC plasma source. Contrary to gliding arc reactors [1], an APGD operates in a glow-like regime, at a few tens of mA and a few kV of voltage drop.

2. Description of the experiments

In this work, we present an APGD reactor for CO_2 conversion, in three distinct configurations, designed based on computer simulations.

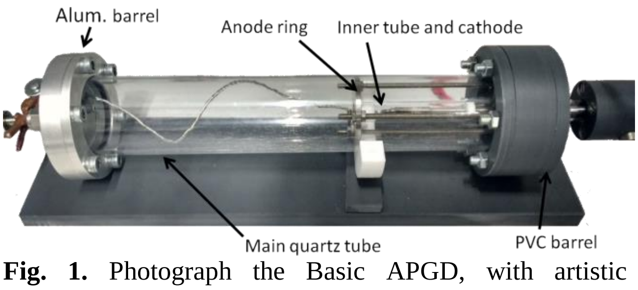


Fig. 1. Photograph the Basic APGD, with artistic representation of the discharge.

The Basic APGD consists of a large quartz tube (300m long), which houses a steel anode ring and a smaller quartz tube for gas delivery (see fig. 1). The cathode is a steel rod with a diameter of 5 mm. The distance between the electrodes is 18 mm. The flow rate is 3 L/min.

The second version of the reactor, the Vortex APGD, is described in figure 2.

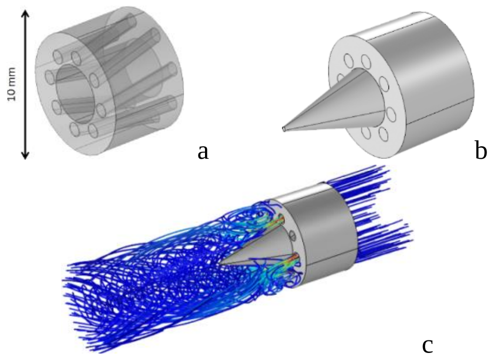


Fig. 2. Vortex APGD – swirl generating brass ring

Utilizing the same body as the Basic configuration, a brass ring with specifically inclined holes surrounds the cathode. The holes lead the gas stream in the direction towards the cathode tip, where the discharge is situated. In addition, they induce a vortex – motion flow pattern. The motivation behind is to (1) cool down the gas with increased turbulence, (2) cool down the cathode as the brass ring acts as a radiator, and (3) increase the residence time for the CO_2 molecules in the plasma region. A lower gas temperature is beneficial for energy-efficient conversion, due to limited VT relaxation, so it promotes the role of the vibrational levels, while the increased residence time should further increase the conversion. The cathode cooling permits usage of a current up to 30 mA, and a electrode distance up to 22 mm (hence higher power input, leading to higher conversion as well).

The third, Confined APGD variant, is a complete re-design of the original reactor. The main idea is to limit the discharge volume, and force a more uniform gas treatment – a known problem for such kind of reactors. The design concept is presented in figure 3.

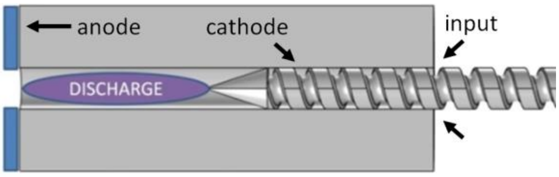


Fig. 3. Schematic of the confined AGPD reactor.

As shown in figure 3, the high-temperature ceramic tube tightly encapsulates the cathode pin. The diameter of the cathode pin is 5 mm, as in the previous designs. A spiral groove is engraved along its body, serving effectively as a gas channel. In this way, two effects are achieved: a vortex motion of the gas, and convective cooling of the cathode. The free volume in the ceramic tube matches the discharge dimensions, i.e. there is no path for the gas to leave the reactor without passing through the plasma. In this configuration, up to 30 mA of current and 1 L/min are possible, leading to a high SEI, and thus a high conversion.

All three reactor configurations are powered with a high-voltage power supply (30kV, up to 40 mA). The gas composition after leaving the reactor is analysed by means of gas chromatography.

3. Modelling methods

The discharge model is developed with Comsol Multiphysics 5.3, using the same approach as in [2]. First, the flow pattern is computed in 3D, yielding a stationary output. The plasma model only permits a 2D geometry, as the computational requirement is too large. Hence, only the Basic APGD could be modelled in detail, since the effects of vortex flow motion cannot be accounted for in 2D. An overview of the boundary conditions is shown in figure 4.

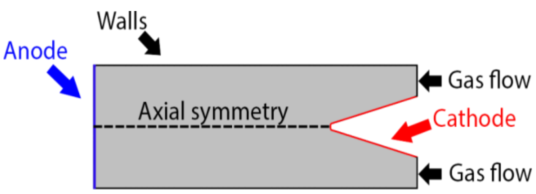


Fig. 4. Boundary conditions for the plasma model.

4. Results

In figure 5, the experimental results are presented, showing the performance of the three reactor configurations.

The Basic APGD yields a conversion around 3.5-4.5 %, for an inter-electrode distance of 18 mm. The Vortex APGD can afford higher current, and in addition, a longer electrode distance, which leads to a significantly higher power input. It shows up to 8.3 % conversion, a significant improvement.

The Confined APGD shows an even higher conversion up to 12.5 % at 30 mA. This is a 50% improvement over the Vortex variant. However, it comes with a price: the energy efficiency is slightly reduced. We attribute this effect to the heat losses due to the contact of the plasma with the ceramic walls.

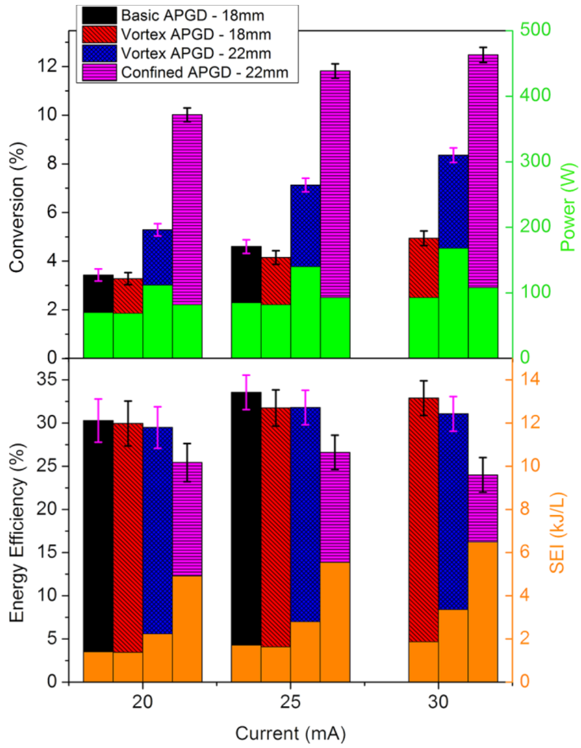
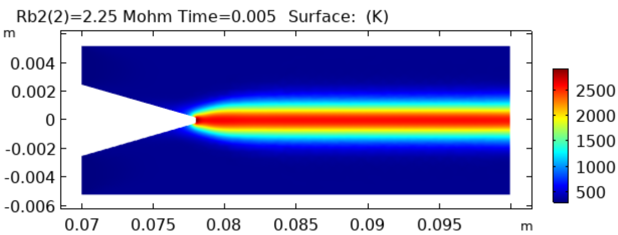
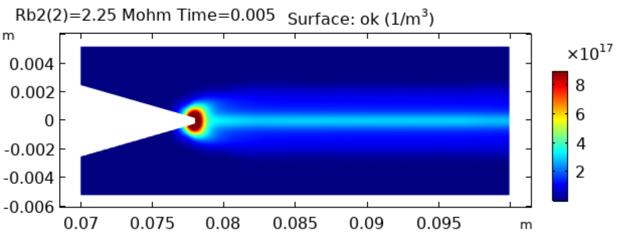


Fig. 5. Summary of the experimental results – conversion, energy efficiency, SEI and power for different discharge currents.

In figure 6, we show the plasma gas temperature, along with the obtained density. The value of 2500 K is in the typical range for discharges at similar conditions [3], and much lower compared to a GAP [2]. Low temperatures are typically more favourable, in terms of conversion efficiency, as they give a stronger thermal non-equilibrium between the vibrational population and the gas.



a



b

Fig.6. Gas temperature distribution obtained from the model (a), and plasma density (b).

5. References

[1] A Fridman, *Plasma Chemistry*, Cambridge University Press, New York, US, 2008

[2] G Trenchev, St Kolev, W Wang, M Ramakers and A Bogaerts, CO_2 Conversion in a Gliding Arc Plasmatron: Multidimensional Modeling for Improved Efficiency, *J. Phys. Chem C*, **121**(44), 24470-24479, 2017

[3] D Staack, B Farouk, A F Gutsol and A A Fridman, Spectroscopic studies and rotational and vibrational temperature measurements of atmospheric pressure normal glow plasma discharges in air, *Plasma Sources Sci. Technol.*, **15**, 2006