

Design of a Lab Reactor Setup for the Study of Radio Frequency Plasma Catalysis of Ammonia

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Introduction and objectives

Ammonia (NH_3) production is one of the key applications of hydrogen process technology. NH_3 is a base chemical with annual production volumes of around 150 million metric tons. It is mainly used as a feedstock for the fertilizer industry, but also for the synthesis of other chemical compounds such as in the textile and pharmaceutical industries and as a potential fuel. Conventional NH_3 production through the Haber-Bosch (HB) process ($\text{N}_2 + 3\text{H}_2 = 2\text{NH}_3$) accounts for 1-2% of the world's total primary energy demand and greenhouse gas emissions (Bicera 2016). A more sustainable ammonia production route uses renewable electricity to not only generate green hydrogen via water electrolysis, but also to assist the HB process. Both electrolysis and electrified HB process are highly electricity intensive, though, and novel electro-technologies are needed to reduce the overall energy needs of the all-electric NH_3 synthesis process. In this regard, plasma-catalysis is an emerging field, in which the plasma excites the gas species (i.e., nitrogen triple bond), which are further converted over the catalyst surface with a lower activation energy. Plasma not only affects the gas species (ionized, excited, electrons), but also influences the catalyst surface (metal dispersion, oxidation state) resulting in possible synergistic effects. Plasma-assisted catalytic conversion of hydrogen and nitrogen to ammonia has been investigated in academic circles for some years now (Li 2018; Carreon 2019, Touvelle 1987, Shah 2018, Peng 2019, Van Helden 2007). Radio frequency discharge plasma promises high conversion rates, but it is still very much in the development stage with limited understanding of kinetics and reaction mechanisms. For the creation of strongly non-thermal plasmas, RF sources are also typically limited to low pressure applications.

The goal of this research at TUM's Chair of Energy Systems is, thus, to design a first lab reactor setup in order to study the plasma catalysis of ammonia for radiofrequency plasma under low pressure conditions. Further, there are two long-term goals. We want to analyze the impact of a wide range of different operating conditions on the synthesis process. In addition, we want to create a second iteration of the lab set up by adding a nano-pulsed DC plasma source to perform combined pulsed-RF plasma catalysis at ambient pressures.



Fig. 2: Result of initial very low temperature ashing experiment for coke sample

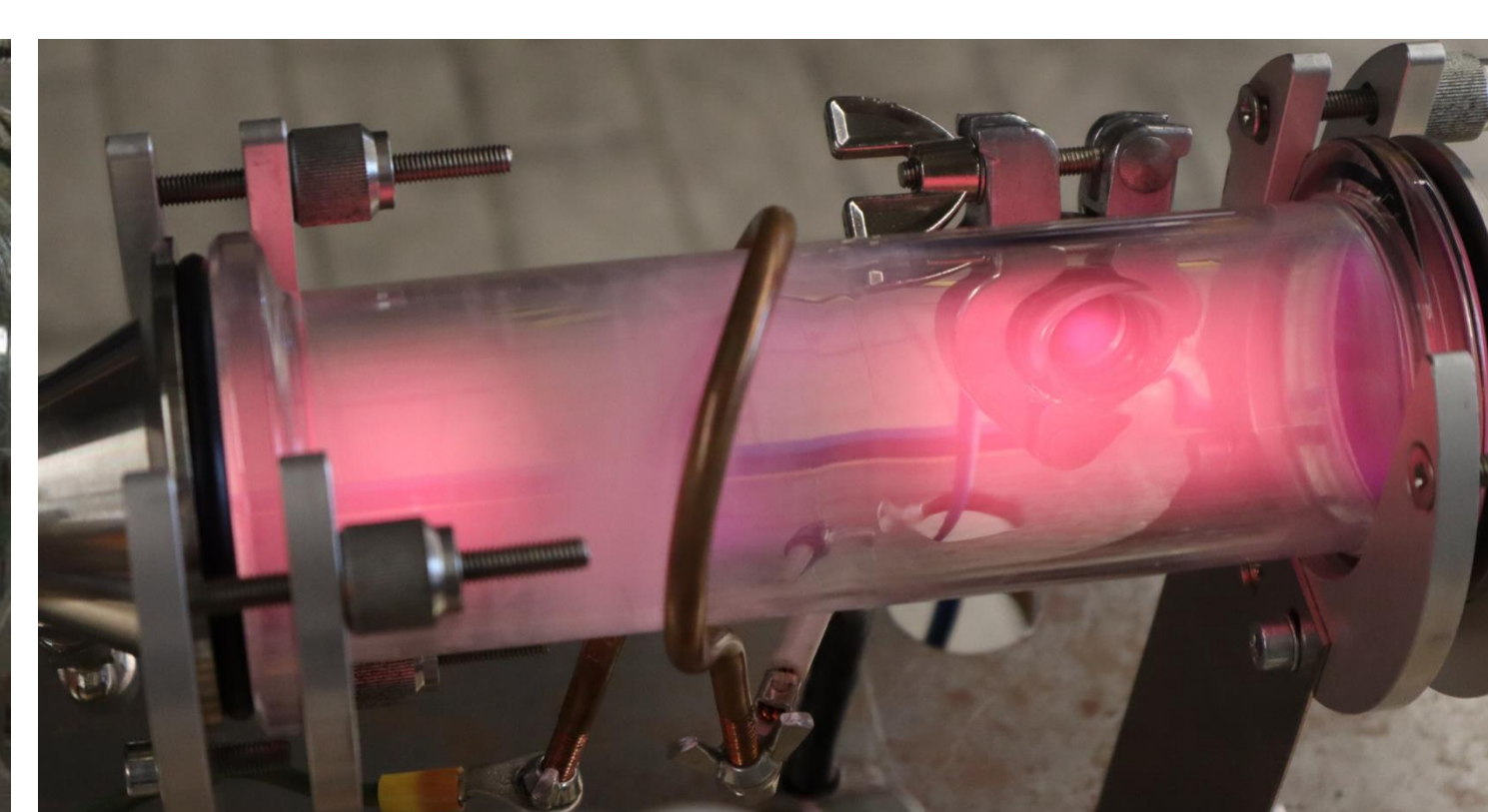
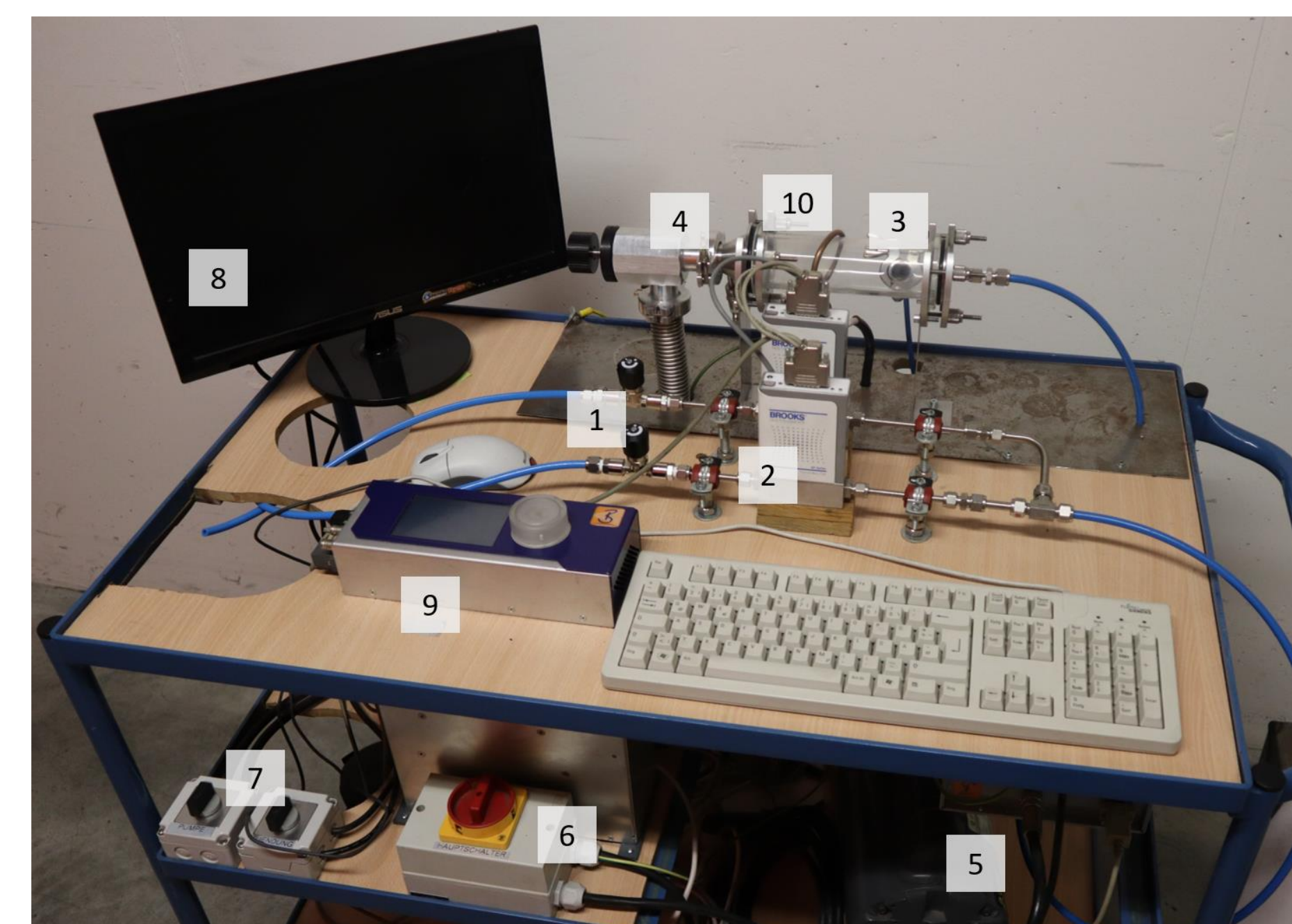


Fig. 3: Non-thermal plasma reactor during operation (protective cover removed)

Base reactor setup

The current reactor setup has been originally designed for very low temperature ashing (VLTA) of biomass and coal samples with the goal of retaining volatile components in the ash phase at operating temperatures below 70°C . The different main components of the lab setup are shown in Fig. 1. At the heart of the lab setup is a low pressure glass reactor with an inductively coupled radio frequency plasma generator. The plasma power can be varied from 0 to 300 W. A matching cube network based on air-dielectric variable capacitors is used to minimize the reflective power of the radio frequency setup. The functionality of the reactor setup has been proven through experiments on very low temperature ashing of coke samples (Fig. 2) and the creation of a non-thermal plasma in nitrogen (Fig. 3).



- 1 – Ball valve
- 2 – MFC
- 3 – Glass reactor
- 4 – Outlet valve
- 5 – Vacuum pump
- 6 – Main switch
- 7 – Pump switch
- 8 – Computer
- 9 – RF generator
- 10 – RF coil

Fig. 1: Overview of main components of radiofrequency plasma lab setup (with protective cover removed)

Modifications for plasma catalysis of ammonia

For the low pressure plasma catalysis of ammonia at sub-atmospheric pressures, we propose to only slightly modify the existing lab setup by adding a metal wool catalyst to the glass reactor as well as a means to heat the glass reactor for achieving catalyst activation. The heat needs to be provided in a way that does not interfere significantly with the RF plasma. Therefore, as a first attempt, a heating oil based electrically insulated heat exchanger will be used. This setup is depicted in Fig. 4.

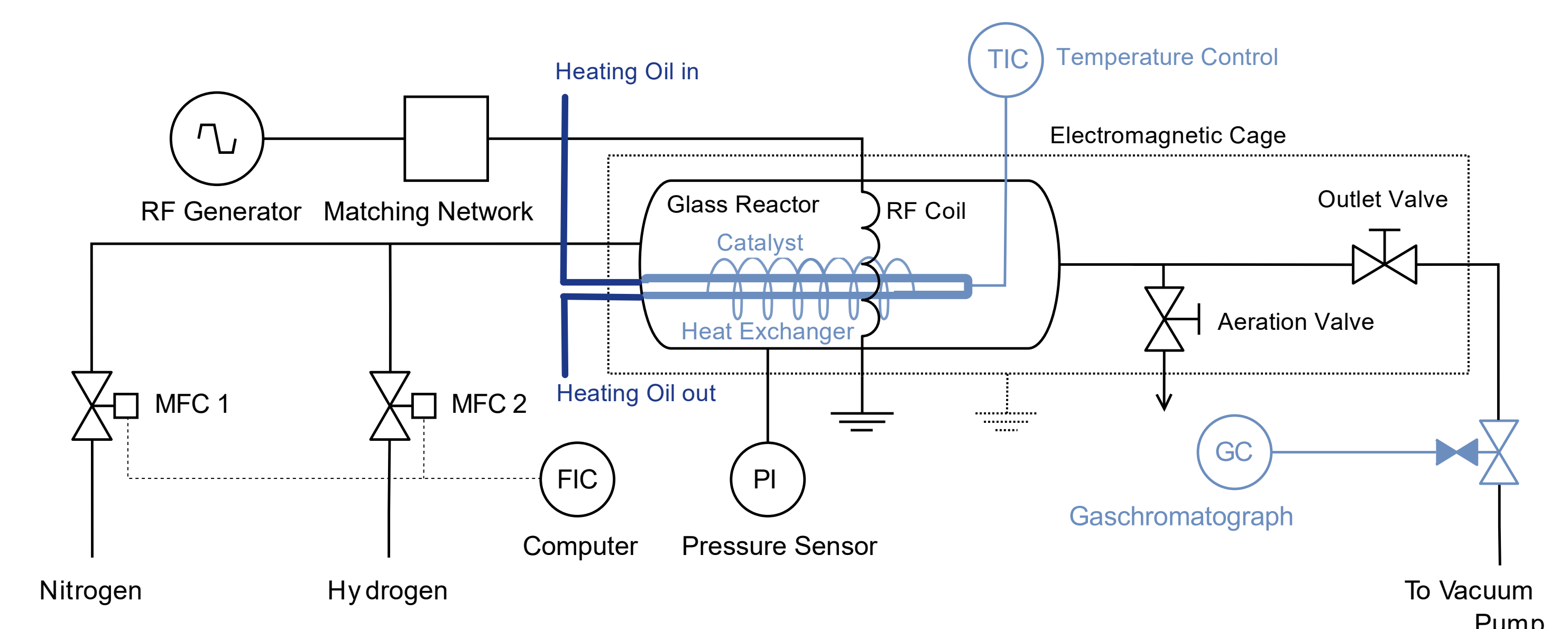


Fig. 4: Design of test rig for radio frequency plasma catalysis of ammonia at low pressures

Next steps

As the immediate next step, first ammonia synthesis experiments will be run at a fixed flow rate for a set period of time, measuring conversion, product yield and energy yield for a given set of operating conditions (H_2/N_2 ratio, temperature, pressure, catalyst). In addition, we plan to install a nano-pulsed DC plasma source to be able to ignite and sustain an RF plasma at atmospheric pressure for ammonia synthesis.

References

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