Experience with operation of a rotating bed reactor pilot for chemical looping combustion (CLC) of methane

Silje Fosse HÅKONSEN, Ivar M. DAHL, Aud I. SPJELKAVIK and Richard BLOM

SINTEF Materials & Chemistry, Norway

Richard.Blom@sintef.no

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In the present contribution we will present the development of a new reactor setup for chemical looping combustion (CLC). The reactor is best described as a rotating bed reactor where the oxygen carrier material is rotated between different gas streams flowing radially outwards through the metal oxide bed. We believe a radially directed gas flow will give the best gas flow performance since volume expansion due to the increase in moles of gas and gas temperature due to the exothermic nature of the total reaction taking place best will be compensated by the geometrical radial reactor volume increase.[1]

A schematic drawing of the rotating reactor is shown in Figure 1. The light blue arrows indicate the radial gas flow direction. The inner black solid lines indicate the separation walls between the various gas inlet sectors (in light blue). From the inlet sectors the gas flows radially through a slowly rotating oxygen carrier bed (the green area) and into one of the two exit chambers (the two outer white chambers with two division walls). The composition of the exit gas from each side of the reactor is then analyzed, by GC, MS and/or IR analyses.

The methane fed through the fuel sector will be oxidized by the oxygen carrier material (in our case CuO/Al_2O_3) producing CO_2 and H_2O, while the air fed on the air side will leave the bed as oxygen depleted air since a certain fraction of the oxygen has been used to re-oxidize the oxygen carrier. The main challenge is to design a rotating reactor where the mixing of the gases on the fuel and air side is as small as possible using a minimum amount of steam in the two sectors in between.

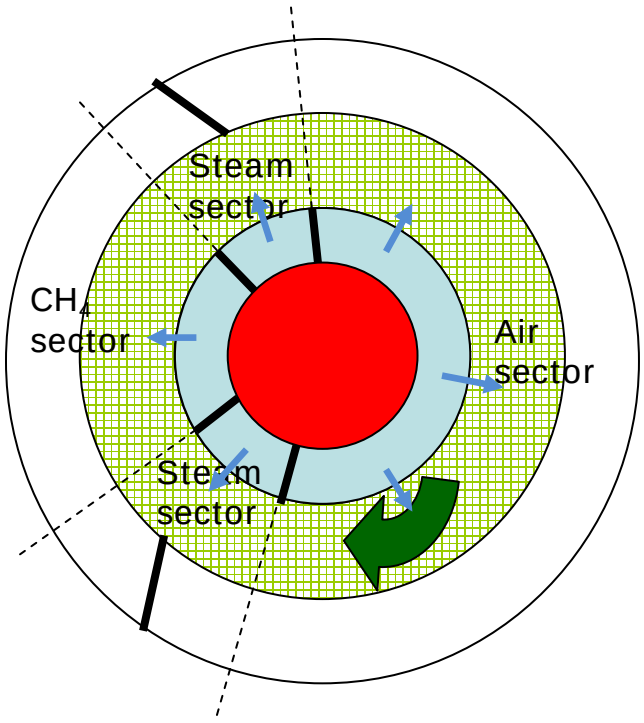

Figure 1: Schematic drawing of the rotating reactor used: The blue part indicate the various gas inlet sectors, the green part if the rotating oxygen carrier bed (viewed along the rotating axis), while the white part show the two exit chambers.

[1] I. M. Dahl, E. Bakken, Y. Larring, A. I. Spjelkavik, S. F. Håkonsen, R. Blom, *Energy Procedia*, **2009**, *1*, 1513.

We have during the last couple of years designed and constructed a small pilot reactor using the principles described above. The reactor has been tested with methane as fuel at temperatures from 600- 800°C using different rotation frequencies (from 0.5- 2.0 rotations/min), different gas feeding rates (total flows between 0.75 and 2.0 l/min), different relative gas feeding rates between the different sectors, etc. Figure 2 show the results obtained when increasing the reactor temperature from 600 to 750°C:

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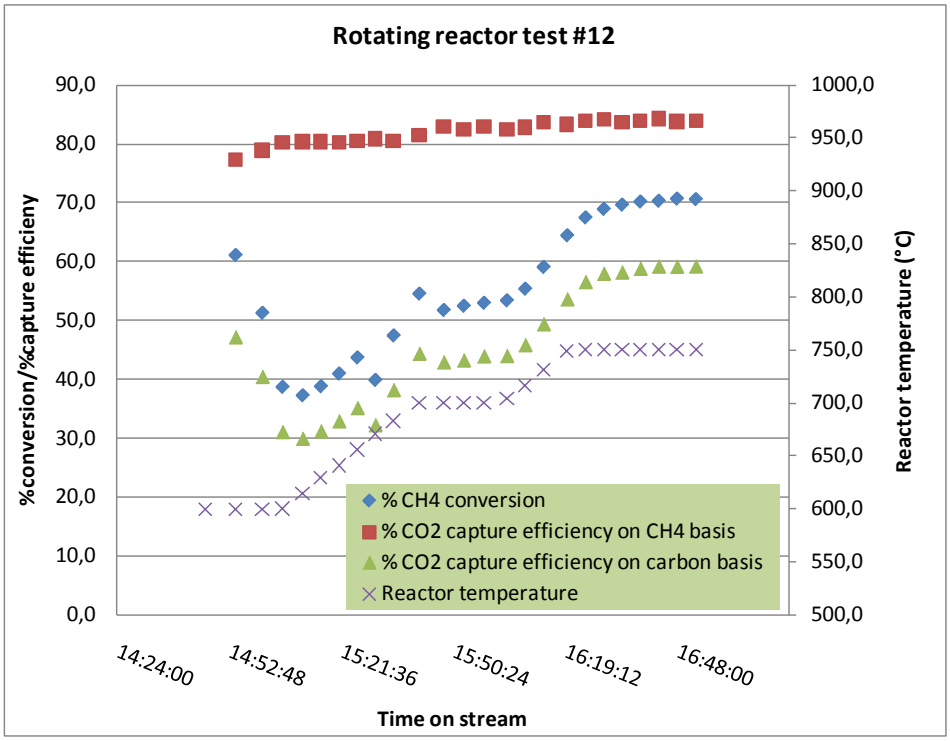

Figure 2: Derived CH_4 conversion and $\%CO_2$ capture efficiency during experiment #12.

Increasing the reactor temperature leads to increased methane conversion and at 750°C 70% CH_4 conversion is obtained. The sole reaction products are CO_2 and H_2O, and no CO was observed at the conditions used. There is also a slight increase in CO_2 capture rate (based on converted methane) going from around 80% at 600°C to 84% at 750°C.

Some gas mixing between the air and fuel side is observed leading to low CO_2 purity (max 70%). The observed gas mixing is mainly a consequence of the specific design choices made for the first reactor version, and we believe significant improvements can be obtained in a second version by using the experience gained for the prototype. In addition, the challenges connected to gas leakage will be drastically reduced when going to large scale reactors with longer relative diffusion distances. At the end of the presentation some aspects with large scale versions of the rotating reactor concept will be discussed.