PULSED COMPRESSION REACTOR CONCEPT FOR SYNTHESIS GAS PRODUCTION

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Introduction

Production of synthesis gas by steam reforming or by partial oxidation involves rather high temperatures of 800 – 1400 ^{o}C. The required heating of the feed and recovering of thermal energy of the product entail high capital and operating costs. These costs are associated with reactors, heat exchangers, furnaces, compressors, heat recovery boilers and turbines, which are made of very expansive high-temperature, scale-resistant alloys that can resist corrosion and withstand the effects of elevated temperatures and pressures. As a result the available synthesis gas based processes for the production of synthetic fuels are currently not economically attractive (Lange and Tijm, 1996).

A fundamentally new chemical reactor concept, namely the free piston pulsed compression reactor (Glouchenkov 1997, 1999), permits a breakthrough in synthesis gas production in terms of energy efficiency, capital costs, and portability. The novel reactor principle is totally opposite to the current trends in improvement of the existing technologies. Instead of developing better catalysts and decreasing the process temperature, in this new reactor no catalysts is required and the reactions occur at very high temperatures of 1500 – 3000 ^{o}C. Via integration of all process steps in a single unit significant improvements in the reactor performance are achieved.

Reactor concept

The basic idea of the reactor is schematically shown in Figure 1. The reactor consists of a double-ended cylinder and a free piston, which divides the cylinder into two compression-reaction chambers. The cylinder has inlet and outlet ports in its wall for feeding the reactants and exhaust of the reaction products. The free piston reciprocates with a very high frequency (up to 400 Hz) compressing in turn the feed gas, until it reacts, in the lower and upper chamber. The reciprocation is maintained by the reaction itself in case of exothermic reactions. An essential feature of the reactor is that the piston-cylinder assembly has no sealing rings. Gas leakage through the annular piston-cylinder clearance is prevented by using contactless labyrinth seals. A relatively small gas leakage is even desired to provide gas lubrication (gas bearings), i.e to prevent any contact of the piston with the cylinder wall by means of self-centering and self-alignment of the piston in the cylinder.

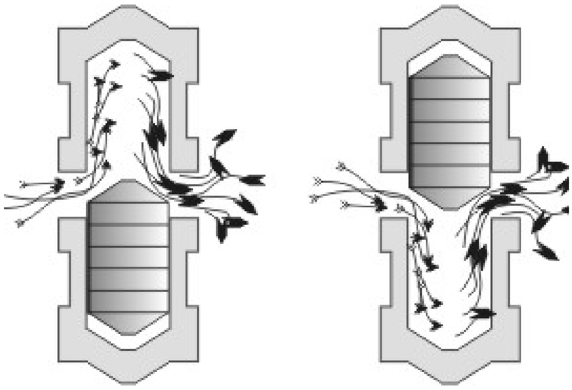

Figure 1. Operating principle of the pulsed compression reactor.

The use of the free, gas lubricated piston, makes it possible to achieve very high compression ratios (the ratios of the volumes before and after compression) and frequencies of the piston oscillation. The resulted very short duration of the extreme conditions prevents a significant heat exchange between the hot, compressed gas and the reactor walls. Therefore unique combinations of pressures and temperatures from several hundreds to several thousands of bars and up to several thousands of K can be obtained. These conditions are far beyond the feasible operation window in steady state chemical reactors. The achieved pressures and temperatures are ideal for almost instantaneous completion of many industrially important chemical reactions. The high frequency of the piston oscillation results in very high space velocities, i.e. ratio of volume throughput to reactor volume (millions per hour). Huge rates of temperature and pressure change (up to 10^7 K/s, 10^7 bar/s) afford an excellent way of "freezing" the high temperature products and producing a better yield in case of equilibrium reactions. Gas compression in the reactor can be adjusted depending on the desired conditions and is not determined by the mechanical restrictions as in conventional piston devices.

The free piston behaves like a pendulum swinging between two gas springs. Compensation of the inevitable energy losses due to friction and gas leakage is only required. These energy losses are very much smaller than the losses in the conventional processes. The reactor comprises the entire processing train: gas compression, heating of the reactants, reaction itself, cooling of products and utilization of the released reaction energy. The reactor is ideally suited for synthesis gas production by partial oxidation of various hydrocarbon feedstocks. Furthermore, the pulsed compression reactor technology may be very interesting for conducting of a great variety of other industrially important chemical reactions, as was proven experimentally using single-shot ballistic piston machines (Ryabinin, 1961).

Experiments and results

Experiments without reactions. Firstly, the basic reactor performance was studied without chemical reactions using two reactors of 105 mm and 70 mm height, both with an inside diameter of 60 mm and many pistons of different density, dimensions and shape. In these experiments, compressed air was injected into the lower chambers either continuously through developed throttles or momentary using developed fast acting valves.

The experiments showed that the reactors can easily be started-up using either of the two start up systems and operate smoothly without wear of the piston and cylinder walls. The reactors demonstrated a unique performance in terms of the achieved combinations of compression, frequency, temperature and pressure: compression ratio 5 – 45, piston frequency 50 – 200 Hz; piston speed 5 – 30 m/s; piston acceleration $(1 – 12)\times10^3$ g. The maximum pressure and temperature observed in these conditions were 200 bar and 1360 K respectively. Such combinations are unique even for the state-of-the-art internal combustion engines and other known piston-compression machines whereas the values of these parameters can easily be increased several times in the developed reactors (Glouchenkov et al., 2002).

Experiments with reactions. To demonstrate the technical feasibility of the new reactor concept two reactors for carrying out chemical reactions – one with a single working chamber and a second with two working chambers as shown in Fig 1 - have been designed, manufactured and tested. The inner diameter of the both reactors was 60 mm. Six pistons of different shape and dimension were used for each reactor.

The experiments were conducted with methane/air, propane/air and ethyl ether/air mixtures. In most of the experiments propane/air mixtures were used; concentration of propane in the feed gas varied from 0.8 to 20 vol % (fuel/air equivalence ratio $\phi = 0.2 – 6.0$).

At first the conditions required for the reactions were studied in experiments without gas flow through the reactors. The occurence of the reactions manifested through a significant increase of the maximum pressures (20 – 200 bar) compared to that at similar conditions but only with air.

The measurements showed unique pressures and reaction rates. Figure 2 shows the influence of propane on the pressure change in the single chamber reactor. It demonstrates in particular the capabilities of the pulsed compression reactors: extremely short reaction times (of about 10 μs) and huge rates of pressure and temperature change. None of the available chemical reactors can provide these operating conditions.

Figure 3 shows an example of the pressure change during continuous operation of the reactor with single working chamber.

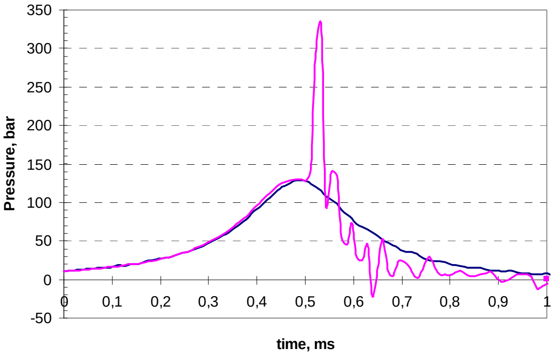

Figure 2. Influence of propane on the pressure change in the single-chamber reactor; dark blue line – without propane, purple line – with propane (ϕ = 0.63).

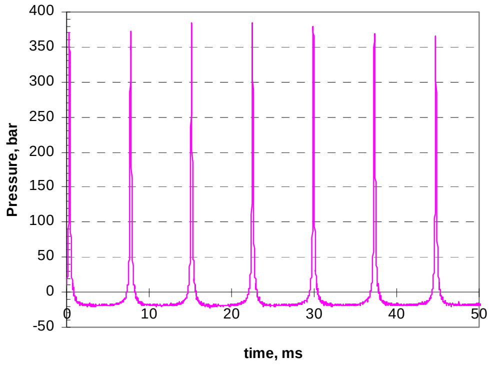

Figure 3. Pressure change in the single-chamber reactor during continuous syngas production: ϕ =3.6.

The analysis of the product composition revealed a significant yield of synthesis gas.

Conclusions

A new free piston pulsed compression reactor was developed. Experiments have shown its feasibility for economic synthesis gas production. Operation of the pulsed compression reactor is optimal in terms of energy efficiency. No other reactors integrate so many functions and allow so high pressures and temperatures, so high quenching rates and so high space velocities.

Furthemore, additional experiments have shown that the reactor permits reaction of propane and oxygen in a very wide concentration range (beyond the inflammability range). This finding may reveal new processes for e.g. manufacturing of carbon black, carbon-based nanoparticles (nanodiamands, fullerens) or direct oxidation of natural gas to formaldehyde, methanol, and higher alcohols.

References

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