PYROLYSIS OF COAL IN A HYDROGEN PLASMA

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ABSTRACT

In order to test the feasibility of direct conversion of coal to acetylene Bergbau-Forschung installed a 30 kW arc heated reactor. Some results from the numerous experiments especially with coals of different rank and different size form < 100 µm to < 5 µm are reported. The dependence of the maximum yield of acetylene from the reaction conditions and from the rank of coal is shown.

1. INTRODUCTION

Mainly gasification and hydrogenation processes have been taken into consideration for the conversion of coal into chemical products. However, a third process line seems to be equally promising, i.e. the conversion of coal into acetylene and the production of chemical feedstocks via this pre-product. The direct conversion of coal to acetylene in the electric arc or plasma reactor was studied in the past on numerous occasions on a laboratory scale; however these studies were merely of academic interest until 1970. In 1971/72, the AVCO-Corporation published data, which appeared to be interesting from an economical and technical point of view.
Acetylene concentrations of 14 % to 16 % in the product gas, an energy consumption of 9 to 11 kWh per kg of acetylene and an acetylene yield of 0,3 kg per kg of coal were reported (1). Bergbau-Forschung has adopted these data in order to compare in a study on economical efficency the production of acetylene in Germany according to this process with the process via calcium carbide and finally with the production of acetylene from hydrocarbons in the electric arc reactor.

2. TEST SET-UP

The thermodynamic and kinetic considerations show the requirements to be made upon a reactor for the production of acetylene from coal or hydrocarbons: the feedstock must be heated very fast to high temperatures and abruptly be chilled after a retention time of a few milliseconds. Fig. 1 gives a schematic view of a plasma reactor which can satisfy these requirements: a mixture of hydrogen and about 10 % argon is heated by a d.c.-arc stabilized in a magnetic field. Hydrogen was chosen since it favours the formation of acetylene. Argon serves only as an inherent standard for gas analysis. The hot gas stream is charged with finely ground coal or with the hydrocarbon by means of hydrogen as a carrier gas. Cold hydrogen is blown into the reactor through the quenching lance to chill the products. The retention time can be varied by displacement of the quenching lance. The acetylene concentration is determined by means of an infrared measuring device. Furthermore, a quadrupole mass spectrometer and a gas chromatograph are available for gas analysis.

3. TEST RESULTS

The temperature of the plasma is an important test parameter; it can be adjusted either by variation of the electric output of the arc or by the dosing speed of the gas streams. Besides, the reaction process is determined by the concentration of the dusty coal in the plasma gas and by the retention time of this coal in the hot zone. The retention time can be influenced by changing the flow velocity of the hydrogen/coal mixture or by altering the hydrogen quenching process, i.e. by altering the quenching point and the amount of hydrogen used. Important test parameters are finally type and grain size of the coal.

For comparative testing to explain the influence of the coal rank, the quenching point was not varied so that only relative values are important in this context (2). It is shown that the gas and acetylene yield related to volatile matter drop essentially with decreasing rank. While the gas yields are in the range of 250 % and roughly 150 %, the acetylene yields drop from about 180 to about 50 % in the coal rank series. This means that under the conditions of the plasma reactor - i. e. extremely high temperature, very high heating velocity and presence of hydrogen - an essentially larger gas volume is generated than under standard conditions, under which the volatile matter content of coals is normally determined.

This is also known from other pyrolysis tests for which it was assumed among others that tars and liquid hydrocarbons are hydrogenated to a large extent due to surplus hydrogen. The modest acetylene yield of brown coal can among others be explained in that the carbon is converted into CO instead of C_2H_2 due to the oxygen present in the coal, as shown by thermodynamic calculation.

As shall be shown in detail, the acetylene yield can be stepped up if the grain size and thus the heating velocity of the coal grain is coordinated with the retention time of the coal in the hot plasma zone. Fig. 2 shows acetylene, carbon monoxide, ethylene and methane yields with quenching at 10, 15, 25, 35 and 50 cm reactor length respectively (3). It gets obvious that in particular the acetylene concentration and thus the acetylene yield is influenced by the quenching conditions. It rises first and passes a maximum level. The other components are practically not influenced by the quenching conditions. The abscissa plots retention time apart from pipe length. The retention time can be calculated according to an idealized model which considers the axial temperature profile of the gases and indicates the flow velocity and thus the retention time up to each point in the reactor via the pressure and temperatur-corrected gas volume at a known pipe section.

Systematic tests with 5 available grain sizes have been conducted. The medium grain diameter is 1 µm for the most finely ground fraction, and 180 µm for the coarsest fraction. The outer surfaces were determined according to the Blaine test and are in the range of 3.4 m^2/cm^3 and 0.3 m^2/cm^3. It was noted that the maximum point shifts to shorter retention times with smaller grain sizes.

The dependence of the retention time at yield maximum on the grain size can be formulated quantitatively, as shown in <u>Fig.3</u>, if the measured Blaine surfaces are used. The retention time required for a maximum acetylene yield is plotted as a function of the Blaine surface which is the larger, the smaller the grain size. In the log-log coordinate system, this results in a straight line with a gradient of - 1/3.

Consequently, the retention time at maximum acetylene yield is proportional to the reciprocal value of the cubic root of the outer surface for the tests described in this paper.

4. DISCUSSION OF TEST RESULTS

Testing of various hard coals has demonstrated that young hard coals with the lowest possible oxygen content allow maximum C_2H_2 yield. Furthermore, the tests have shown that the acetylene concentration is higher than the thermodynamically calculated balanced state concentration. Finally, a correlation between the retention time for maximum C_2H_2 yield and grain size of the coal was established.

For an interpretation of these results it must be considered that the heating velocity depends on the grain size of the coal, and furthermore that the velocity with which the volatile pyrolysis products are formed is the higher, the higher the heating velocity. The reactions expected for coal in the electric arc plasma are schematically shown in <u>Fig. 4</u>. The first step is the pyrolysis of the coal linked up with the diffusion of the pyrolysis products from the coal grain. The gaseous pyrolysis products, which mainly consist of hydrocarbons, e.g. methane and radicals, disintegrate to soot and hydrogen, whereby acetylene is generated as an intermediary product. Parallel to this, a hydrogenating coal gasification may also take place, e.g. by means of the atomic hydrogen present in higher concentrations in the electric arc plasma.

The gasification products are again hydrocarbons and radicals which may react to acetylene.

Since acetylene does not arise as a stable end product of the reaction but as an intermediary product, the acetylene concentration passes a maximum level similar to the methane disintegration process at high temperatures. The acetylene yield is highest when the gas mixture is chilled at the time the maximum level is reached. The retention times are in the range of few milliseconds.

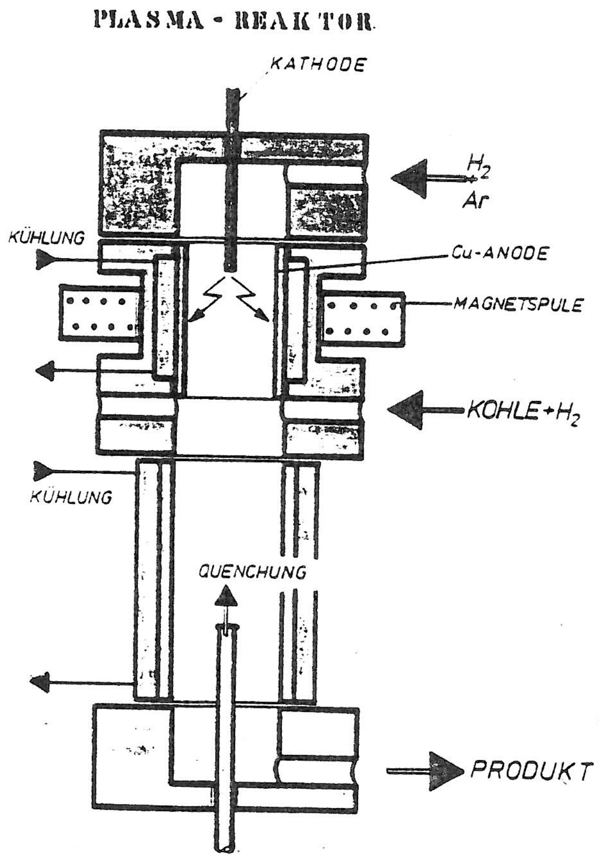

Fig.1. Plasma reactor

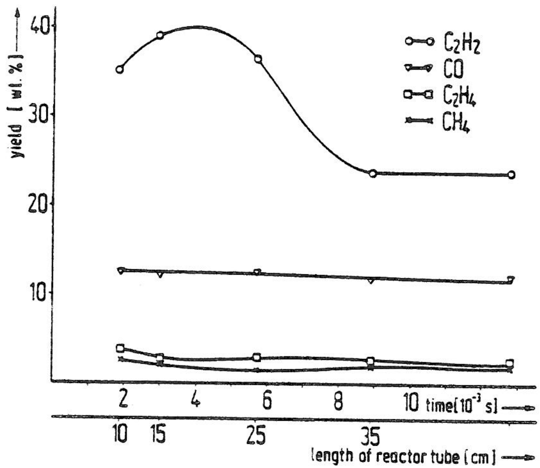

Fig.2. Product gas yield resulting from conversion of long flame gas coal with quenching.

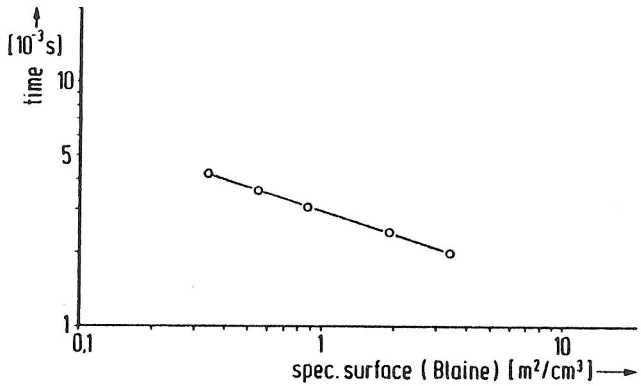

Fig.3: Retention time at maximum level of acetylene yield as a function of Blaine surface

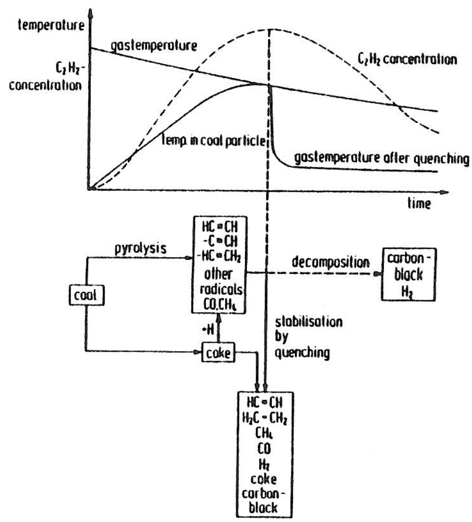

Fig.4: Schematic diagram of high-temperature pyrolysis reaction

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