

The invention relates to a method for indirectly heating a process gas stream in a reaction space for an endothermic, in particular catalytic reaction according to the preamble of claim 1 and to a device for carrying out this method.

Catalytic steam reforming of hydrocarbons to produce a gas which is rich in hydrogen represents a possible field of application of the invention. A mixture of water vapour, a hydrocarbon and/or carbon dioxide is used as the process gas. This gas mixture is supplied at temperatures of approximately 400 to 600°C and pressures of up to 4 MPa to a reaction space, which may consist of a plurality of catalyst-filled tubes, where it is heated to approximately 750 to 900°C. This gas mixture then reacts endothermically over the catalyst to produce a gas which is rich in hydrogen and contains carbon monoxide and carbon dioxide and excess water vapour and hydrocarbon.

The hydrogen content of the generated product gas depends, inter alia, on the excess of the water vapour which is used and on the temperature and the pressure in the catalyst-filled tube. An increase in the water vapour excess and the temperature in the catalyst-filled tube gives rise to an increase in the generation of hydrogen, while a pressure increase results in a decrease of the latter.

The EP 0 194 067 B1 discloses a device for carrying out an endothermic catalytic reaction of a process gas stream. This comprises a housing with one or more catalyst-filled tubes which are arranged parallel to one another in the housing and each of which has one blind end. The housing comprises means for supplying and removing a burnt gas for heating the outer surface of the catalyst-filled tubes, through which the process gas stream is conveyed while absorbing heat. The catalyst-filled tubes are each surrounded by a tubular shell which forms an annular gap with the tube. The shell extends over the greater part of the length of the catalyst-filled tube. The inlet for the burnt gas is directed directly at the blind end of the catalyst-filled tube. The hot burnt gas

consequently flows directly against the catalyst-filled tube. This has proved to be disadvantageous, as it gives rise to a very high thermal load on the blind end of the catalyst-filled tube. The temperatures produced at critical points in the shell of the catalyst-filled tube are so high that it is damaged after a relatively short time and has to be replaced if the burnt gas is not appropriately cooled beforehand. This could, for example, be carried out by a series-arranged heat exchanger or by mixing with burnt gas which has already been cooled. Both measures entail corresponding additional expenditure on construction work.

In order to moderate the thermal load on the catalyst-filled tubes, it is known to provide the outside of the latter with a ceramic protective casing in the area in which they are directly acted upon by the hot burnt gas. A disadvantage of protective casings of this kind lies in the fact that they are susceptible to thermal expansion stresses which cause damage to the ceramic material and then, after a period of time, to the actual catalyst-filled tubes.

In order to solve the problem of the walls of a reaction space overheating, it is proposed in the applicants' patent application EP-A 369 556, which is not a prior publication, to initially convey the hot burnt gas used to heat the reaction space walls along a barrier of a heat-resistant and efficiently heat-conducting material and in so doing let heat be delivered to this barrier until the gas reaches a temperature which is harmless to the reaction space walls. Only then is the burnt gas stream deflected and conveyed further in the opposite direction on the other side of the barrier and, having already cooled slightly, it comes into direct contact with the walls of the reaction space and delivers heat to these walls. Since, however, the burnt gas simultaneously absorbs heat from the heated barrier, its temperature is maintained almost constant, in spite of delivering heat to the reaction space in the area of the barrier, which only extends over a part of the length of the reaction space. The burnt gas cools relatively quickly

outside of the barrier area as it travels along the reaction space, so that the temperature in the reaction space has a corresponding gradient.

Whereas it is only damage to the walls of the reaction space due to heat which is to be prevented in the EP-A 369 556, the object of the present invention is to optimise the thermal conditions for an endothermic reaction, in particular a catalytic reaction, such that isothermal conditions are largely present throughout the reaction space.

This means that the reaction space should not have a steep temperature gradient, with the desired endothermic reaction taking place in the different areas with correspondingly different degrees of intensity, as has been the case until now, but that an improved utilisation of the reaction space should be achieved by creating reaction conditions which are approximately equally satisfactory, from the thermal aspect, almost everywhere.

This object is solved by a method having the features of claim 1 and is advantageously developed by the characterising features of claim 2. A device for carrying out this method is indicated in claim 3. The characterising features of the subclaims 4 to 11 represent advantageous developments of the device according to the invention.

The invention utilises the knowledge from the EP-A 369 556 and broadens the latter's field of use substantially by ensuring that the indirect heat exchange between the flow sections of the same burnt gas stream which are conveyed in opposite directions along opposite sides of a heat-conducting barrier is maintained over the entire axial length of the reaction space. This enables the delivery of heat in the burnt gas flow section which faces the walls of the reaction space to be partly compensated by the absorption of heat which can take place through the heat-conducting barrier from the oppositely directed burnt gas

flow section. The temperature of the burnt gas which is conveyed past the walls of the reaction space in direct contact with them therefore drops comparatively slowly, in spite of the constant delivery of heat, which takes place by convection, to the reaction space.

On the other hand, a heat exchange takes place between the barrier and the walls of the reaction space due to heat radiation. The trend of the intensity of this heat radiation over the axial length of the reaction space is the reverse of that of the burnt gas temperature in the flow section which faces the reaction space walls. As a result, the walls of the reaction space, through which the heat exchange takes place, heat up almost to the same temperature, and a temperature which is approximately of the same level everywhere also develops in the reaction space, in spite of the endothermic reaction processes taking place, which temperature can be set to an optimum level (according to the type of reaction).

For this to take place, however, the process gas stream introduced into the reaction space must already have been preheated approximately to the temperature of reaction. A zone of a lower temperature will of course otherwise result in the reaction space, within which zone the gas is heated to the temperature of reaction. The heat still contained in the cooled burnt gas which has been drawn off can be used to preheat the process gas.

The invention enables a reaction space of a given size to be utilised in the best possible way by maintaining the temperature at a constant maximum value almost everywhere. This maximum value is selected such that the permissible thermal stresses in the materials used for the reactor are not exceeded and the most favourable thermal conditions possible are at the same time provided for the respective chemical reaction.

The invention is described in detail in the following on the

basis of the embodiments illustrated in Figures 1 and 2, in which:

Figure 1 is a reactor according to the invention with a cylindrical cavity in the reaction space,

Figure 2 is a reactor according to the invention with a plurality of cylindrical cavities in the reaction space.

The reactor 5 shown in Figure 1 has a reaction space 1 with an outer cylindrical shell 2. The reaction space 1 is essentially annular, as it comprises an inner cylindrical cavity 3, which is defined by an inner wall 4 of an efficiently heat-conducting material.

A preheated process gas (e.g. ethyl benzene vapours) can be introduced into the reactor 5, which is of the upright type, at the bottom via the inlet 6. This process gas initially flows into an annular distributor space 22, and from the latter upwards into the actual reaction space 1, which is filled with a catalyst, for example. After flowing through the catalyst substance, the process gas is converted into a product gas (e.g. styrene) and leaves the reaction space 1 at the top end 12 of the reactor 5 through the product gas outlet 7. In order to maintain the endothermic reaction in the reaction space 1, the latter must be heated by means of a hot burnt gas. For this purpose a burnt gas inlet 10 is provided at the bottom end 13 of the reactor 5, through which inlet hot burnt gases are conveyed directly into the cylindrical cavity 3. The burnt gas inlet 10 is advantageously formed as a combustion chamber 16 with refractory walls 15, so that the burnt gas can be produced directly in the reactor 5.

For this purpose the combustion chamber 16 comprises at the bottom end a burner 19, to which fuel and a gas containing oxygen can be supplied via the lines 20 and 21. A barrier which is formed as a tubular member 8 from an efficiently heat-conducting material directly adjoins the burnt gas

inlet 10 inside the cavity 3. This gastight tubular member 8, which extends almost to the top end of the cylindrical cavity 3, in the first place protects the inner wall 4 of the reaction space 1 against direct contact with the hot burnt gas and forms an annular gap 9 with the wall 4. This annular gap 9 is open at the top towards the inside of the cylindrical cavity 3. The hot burnt gas produced by the burner 19 therefore initially flows upwards along the tubular member 8, is deflected outwards at the top end of the tubular member 8 and finally flows downwards in the opposite direction along the outside of the tubular member 8 through the annular gap 9, where it can be drawn off through the burnt gas outlet 11 disposed at the bottom end 13.

The burnt gas flowing downwards through the annular gap 9 constantly delivers heat via the wall 4 to the process gas flowing in counterflow through the reaction space 1 and enables the endothermic reaction to take place in the latter. As hot burnt gas simultaneously flows along the inside of the tubular member 8 and delivers heat to the latter, some of this heat is transferred to the flow section of the burnt gas stream which is flowing downwards, so that its temperature drops comparatively slowly, in spite of the fact that heat is continuously delivered to the process gas.

The extent of the heat exchange between the two flow sections of the burnt gas stream and between the burnt gas stream flowing downwards and the process gas stream can be influenced by the choice of the diameter of the tubular member, as a greater diameter, with conditions otherwise the same, results in a reduction in the size of the annular gap 9 and thus an increase in the flow speed in the downward flow section of the burnt gas, so that, due to the intensified heat exchange (convection) with the inner wall 4 of the reaction space 1, the burnt gas enters the burnt gas outlet 11 at a lower temperature. The heat exchange between the two oppositely directed flow sections of the burnt gas can be influenced particularly effectively by arranging a flow displacement member 14 inside the cylindrical cavity 3

in the area of the latter's longitudinal axis, which member extends approximately over the length of the tubular member 8 and keeps the flow section of the burnt gas which is directed upwards in the vicinity of the inner surface of the tubular member 8. The flow speed of the burnt gas stream which is directed upwards increases in accordance with the reduction in cross section, so that an improved heat exchange through convection is achieved.

The burnt gas flowing upwards out of the combustion chamber 16 along the tubular member 8 causes substantial heating of the wall of the tubular member 8, the temperature of which decreases as the height increases. The radiation of heat from the wall of the tubular member 8 onto the wall 4 of the reaction space 1 is clearly greater in the lower part than in the upper part. This results in an appreciable equalization of the temperature in the wall 4 over the length of the reaction space 1, so that isothermal conditions are also largely established inside the reaction space 1.

In order to facilitate the deflection of the burnt gas stream at the top end of the tubular member 8, the reaction space 1 has an approximately cupola-shaped base 18 internally at the top. Owing to the constant delivery of heat in the area of the burnt gas stream which is directed upwards, the temperature of the latter can be specifically lowered, until the top end of the tubular member 8 and thus the cupola-shaped base 18 is reached, at least to an extent such that direct contact between the burnt gas and the mechanically loaded inner wall 4 of the reaction space 1 does not result in thermal damage.

The reactor 5 may be provided on the outside with a heat insulation 17, which reduces the radiation losses, in order to improve the thermal efficiency. In some cases, in which the burner produces a burnt gas at a very high temperature, it is also advisable for some of the cooled burnt gas, which is drawn off through the burnt gas outlet 11, to be returned

to the burner 19 or the combustion chamber 16, in order to reduce the burnt gas temperature therein. Although the heat exchange between the burnt gas and the process gas is advantageously carried out in a counterflow process, particularly when the process gas is not heated to the temperature of reaction, it is nevertheless also possible to carry out the invention according to the parallel flow principle. The housing is advantageously made of a refractory material in the lower part 13 of the reactor 5, as is the flow displacement member 14, which may be formed as a solid rod or as a tube with at least one closed end. Metallic materials are recommended for the shell 2, the walls 4 and the tubular member 8.

A modified arrangement of the invention is shown in Figure 2, in which parts having the same function have been given the same reference numbers as in Figure 1.

The reactor 5 shown in this figure comprises a plurality of cylindrical cavities 3, which are arranged parallel to one another and the cylindrical walls 8 of which in each case communicate in a gastight manner with the holes of a perforated base 24. The tubular members 8, which are introduced coaxially into the cavities 3 from the bottom, in each case communicate in a gastight manner with the holes of a second perforated base 23, which closes the combustion chamber 16 towards the top, so that the hot burnt gases are forced to rise through the tubular members 8, are deflected at the top ends of the latter in the area of the cupola-shaped bases 18 and conveyed downwards through the annular gaps 9. The heat exchange between the burnt gas flow sections and the process gas takes place in the same manner as in Figure 1. After flowing through the annular gaps 9, the cooled burnt gas collects in the space between the two perforated bases 23 and 24 and then leaves the housing of the reactor 5 at the bottom through the burnt gas outlet 11.

The invention provides, in particular, the following advantages: The reactor is of an extremely simple structure

and therefore entails comparatively low production costs. The heat flow through the wall is uniform and can be approximated to an optimum maximum value which allows fully for the possibility of the materials used being subjected to a thermal load. Problems due to mechanically loaded parts being directly acted upon by an open flame or burnt gases which are too hot are prevented. As the endothermic reaction can take place practically under isothermal conditions, the reaction space volume can be used in a highly efficient manner. Given a predetermined output, the comparatively small structure which is thus possible enables the radiation losses to be minimised. No refractory materials are required for external heat insulation; these are only recommended in the area of the burnt gas inlet and outlet.

Claims:



1. Method for indirectly heating a process gas stream in a reaction space for an endothermic, in particular catalytic reaction, in which a hot burnt gas flows around the walls of the reaction space at the surface which is disposed outside the reaction space and the heat absorbed by the walls is delivered to the process gas, the hot burnt gas initially being conveyed along a barrier, which is formed from a heat-conducting and heatproof material, while delivering heat to this barrier, and, after reaching a temperature which is harmless to the walls of the reaction space, being deflected and conveyed further in the opposite direction on the opposite side of the barrier, after which it is brought into contact with the outside of the walls of the reaction space, and the burnt gas which is conveyed further in the opposite direction absorbing heat from the barrier on one side and delivering heat to the walls of the reaction space on the other, characterised in that the hot burnt gas is conveyed along the barrier over the entire axial length of the reaction space before being deflected and, taking account of the heat exchange between the burnt gas and the reaction space, the heat exchange between the opposite flow sections of the burnt gas, which takes place through the barrier, as well as the heat exchange by heat radiation between the barrier and the walls of the reaction space, the flow speeds of the burnt gas stream in the flow sections before and after the deflection are adapted to one another such that the walls of the reaction space which absorb heat are substantially of the same temperature over the axial length.

2. Method according to claim 1, characterised in that the process gas stream is conveyed in counterflow to the burnt gas stream which is conveyed along the walls of the reaction space.

3. Reactor for carrying out the method according to claim 1, with a reaction space (1), which is enclosed by a cylindrical shell (2) and in which at least one cylindrical



cavity (3) is formed, which cavity extends essentially over the length of the reaction space (1), is directed parallel to the latter's longitudinal axis and is in each case separated from the reaction space (1) by an inner wall (4) of an efficiently heat-conducting material, with a process gas inlet (6), which is arranged at one end (13) of the reactor (5), to the reaction space (1) and a product gas outlet (7), which is arranged at one end (12) of the reactor (5), from the latter's reaction space (1), with a respective tubular member (8), which is coaxial with the cavity or cavities (3) and which forms an annular gap (9) with the inner wall (4) of the reaction space (1), and with a burnt gas inlet (10) and a burnt gas outlet (11), which communicate with the annular gap (9), characterised in that the tubular member (8) in each case extends inside the cylindrical cavity (3) over almost the entire length of the reaction space (1), that the process gas inlet (6) and the product gas outlet (7) are arranged at opposite ends (13, 12) of the reactor (5), that the burnt gas inlet (10) and the burnt gas outlet (11) are arranged at the same end (13) of the reactor (5), that the burnt gas inlet (10) leads directly into the tubular member (8), and that the annular gap (9) is open towards the inside of the tubular member (8) at the end (12) which is opposite the burnt gas inlet (10).

4. Reactor according to claim 3, characterised in that the product gas inlet (6) is arranged at the end (13) of the reactor (5) at which the burnt gas inlet and outlet (10, 11) are disposed.

5. Reactor according to claim 3 or 4, characterised in that a coaxial flow displacement member (14) for the hot burnt gas is arranged inside the tubular member (8).

6. Reactor according to one of claims 3 to 5, characterised in that the inlet (10) for the hot burnt gas is formed as a combustion chamber (16) with refractory walls (15).



7. Reactor according to claim 6, characterised in that the outlet (11) for the cooled burnt gas comprises a conduit connection to the combustion chamber (16), via which some of the burnt gas stream can be returned to the combustion chamber (16).

8. Reactor according to one of claims 3 to 7, characterised in that the reaction space (1) is provided on the outside with a heat insulation (17).

9. Reactor according to one of claims 3 to 8, characterised in that the reactor (5) is of the upright type.

10. Reactor according to claim 9, characterised in that the reactor end (13) with the burnt gas inlet (10) and outlet (11) is arranged at the bottom.

11. Reactor according to one of claims 3 to 10, characterised in that the reaction space (1) has a cupola-shaped wall (18) in the area of the open junction point between the annular gap (9) and the inside of the tubular member (8).