

Reformation of Burnable Gas

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ABSTRACT: New reforming device of an organic fuel that produces a synthesis gas containing CO and H₂ are fabricated. The active part of the reformer consisted of the wall flow manifolds decreases the fuel-air mixture gas speed at the reformation region. The device was applied for the reformation of a propane gas. Reformation of the propane shows up to 71% transforming of carbon into CO, its concentration can reach 18 % of the outgoing gas.

Keywords: gas reformation, synthesis gas, CO production

1. INTRODUCTION

Organic matter has great potential as a clean, renewable feedstock for producing modern energy carriers [1-3]. Many papers on the methane reformation that produces synthesis gas containing hydrogen and CO by a partial oxidation were published [4-7]. The hydrogen in the reformed gas can be used as a fuel in a fuel cell and CO as a reduction agent of a chemical engineering process.

This paper focuses on the process of burnable propane gas reformation by a new device designed for an increase of reforming efficiency. In our studies we are producing a small scale, portable reformer without catalyst, to be less dependent on fuel impurities. But the issue in the catalyst use is the cost as well as contamination of catalyst leading to efficiency reducing and life time decreasing.

2. EXPERIMENTAL EQUIPMENT

2.1 Reformer

The reformer consists of three main parts. The first one is an active element - Si-C filter of Notox ø 20, length 103 mm with an electrically conductive coating Pyro-Duct 597-C of Aremco Products inc. on the cylindrical wall. The second part is a propane and air supply system. The last part is housing with thermal insulation, electrode feeds throw, openings to fixed and movable thermocouples, opening to pressure gauge, opening to propane – air mixture input. The drawing of the system assembly is shown on the Fig 1.

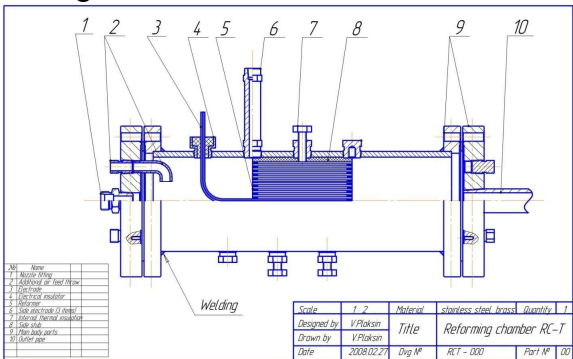


Figure 1: Reforming apparatus. 1 Burnable mixture fitting; 2 Additional air

feeds throw; 3 Electrode; 4 Electrical insulator; 5 Reforming member; 6 Thermocouple fitting; 7 Side electrode (3 items); 8 Internal thermal insulator; 9 Main body parts; 10 Outlet pipe.

2.2 Reforming member

The use of a reformer to convert a liquid fuel into a gaseous fuel stream containing hydrogen and carbon monoxide as major components such as catalytic partial oxidizers (CPOx), non-catalytic (homogeneous) partial oxidizers (POx), or auto thermal reformers (ATR), to generate from engine fuel, water and/or air, streams including hydrogen and carbon monoxide is known. As a reaction area they use chambers, pipes, pellets, powders, foams, honeycombs, fibres. [See for example WO 2001/052978, WO 2001/024922, WO 2001/012540.] In all of these reaction elements the gas flows mostly in axial direction, this is why the reformation rate is limited whether by the flame flow velocity, whether by the contact area between gas and surface, in the last case the backpressure of device is also a subject to consider in practical application. However the efficiency of the reformer by turning of the flame front whereby increasing the effective burning surface, what has been done using the labyrinth structure of reformer in the investigation.

The present work have examined a structure which cannot be seen in the conventional technique, in which the fuel – air mixture flow throw the reforming member is organised in a way to form a burning front of increased area exceeding manifold the cross-section of the reformer. Thus, the increasing of the productivity and possibility to decrease or eliminate the presence of catalyst is has been achieved.

Eliminating of the catalyst or making its role is one of the tasks of the work, because of manufacturing expenses and possible contamination of catalyst with impurities of supplied fuels.

As an active reforming element we directed to a columnar honeycomb structural body comprising a large number of through holes placed in parallel with one another in a length direction with a wall portion interposed there between, wherein: each of the through holes has one of ends sealed;

one end face of the through hole differs in opening area from the other end face thereof;

a ceramic material which constitutes the wall portion is porous.

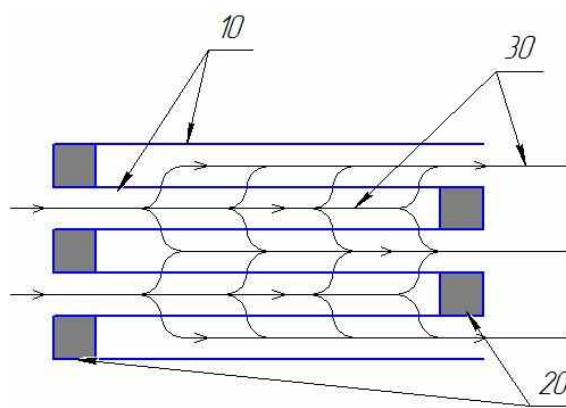


Figure 2: Schematic plan views of reforming member.

The honeycomb structural body with embedded heating system is covered on its side wall with a heat insulation layer, which will prevent heat losses from the reforming member to the reactor tube. In case if the reforming member is heating by electrical current running throw said member the insulation layer must be dielectric if voltage is supplying with two electrodes, if voltage is supplying with one central electrode and another electrode is connected to the tube the insulation can be electrically conductive or conductor should connect reforming member and tube.

The reforming member consist of honeycomb structural body made of silicon carbide, and has a structure in which a plurality of porous ceramic walls 10 (Fig.2) are combined with one another through a sealing material layer that serves as an adhesive to form a ceramic block, and a sealing material layer is also formed on the

circumference of this ceramic block so as to prevent leakage of gases.

Each of the through holes, formed in the porous ceramic member, is sealed at one of ends of its exhaust gas inlet side or outlet side 20 (Fig.2), so that gases that have entered one through hole are discharged from another through hole after having always passed through a partition wall that separates the through holes. The schematic depiction of the fluids flow is marked with number 30 (Fig.2).

2.3 Operation Method

The aim of the device is to perform a controllable burning of the propane – air mixture. This kind of mixture is not always easy to ignite. This is why the ignition must be of volume type, for example discharge or active media. In this invention a honeycomb structural body is used as an igniter.

The operation of the reforming device is performing in three stages: preheating, start-up and reforming. On the first, preheating stage the reforming member is heated at least in part to temperatures about 800 °C with a heater. The heater can be electrical and connected to the honeycomb structural body, or heating can be performed by burning of fuel – air mixture in front of honeycomb structural body started with an embedded igniter.

Next stage – start-up, the fuel–air mixture is switching on or adjusting and it starts to burn at the hot region of preheated reformer. Now the entire reformation region will be heated by a heat released in a reaction of oxidation. The components ratio is established to get the maximum heat from the reaction.

The final stage – reforming. The propane –air mixture is adjusting, the amount and quality of mixture are settled to maintain the proper temperature (800-1500 °C) inside the reformer and maximum yield of products. The ratio of components should be close to stoichiometric ratio for partial oxidation

reaction combined in presence of water with water-gas shift reaction.

The control and switching of operation modes can be carried out using devices for temperature and gas composition measuring embedded into device particularly into honeycomb structural body.

2.4 Effect of the Labyrinth Flow

One of the problems of reformation using the partial oxidation reaction or water – gas shift reaction is the flame. The flame velocity usually doesn't exceed 50 cm/sec. This is why the productivity of reformer is limited by its geometrical sizes or cross-section. To accelerate the reformation they use catalyst or plasma.

In present invention the structure of honeycomb structural body with blocked holes ends forms different burning front. Gas flows not in parallel to the axis as in other reformation devices, but reforming gas flows throw the hole walls and exit the hole wall in direction perpendicular to the hole axis with velocity much lower than in other parts of the device. This happens because the walls gas flow resistance is much higher than the one of the hole, and the flowing gas is looking for the pass throw with lower resistivity, means perpendicular to the ceramic members 10. That means that the front of burning will be as big as the surface of all walls of holes having opening to the outlet. This burning surface is tens or hundreds times greater than the cross-section of reforming member.

The limit for device productivity with absence of catalyst can be estimated in a following way. First the surface S of all walls of holes having opening to the outlet to be estimated. Flame velocity F is usually known for each particular reaction. So the volume V of reactant to be reformed will be not bigger then F times S .

$$V < F \cdot S$$

3. EXPERIMENT

3.1 Procedure

Initially the Si-C block was heated through several steps. First use AC 100 V for about 2 min. until the temperature become 150-160 °C. After this temperature the resistance of the block decreases below 100 Ω. At this moment current limitation become necessary because fixing of the voltage leads to the steep increasing of the current and can cause equipment damage.

On the next stage DC 70 V – 15 A is connected. It starts with 70 V – 1 A voltage limitation mode, and in a few minutes finishes with 20 V – 15 A current limitation mode at temperatures about 230 °C. The last power source to connect was 50 V – 200 A. Using this one we gradually increased current from 15 A to 40 A during 20 min. reaching the temperature about 550 °C.

After the gases supply is switched on – fuel is about 3.3 l/min, air 20 l/min. The heat from chemical reaction works together with DC to heat the block. At 800 °C DC can be turned off to prevent overheating.

It is worth to be noticed that the temperature profile is different in case of current and reaction heating. The power electrode is located in the centre of the block; at temperatures of the side wall below 400 °C the neighbouring four holes are shining indicating temperatures above 800 °C. With increasing of temperature the central part remains the brightest. Burning on the contrary heat all the block radius uniformly, there is not a big difference in the light colour. Low fuel rate can maintain the reaction, but the exit part of the reformer at such flow was not red hot. The red hot was only the inlet of the reformer, and some part of the mixture passes through the region with a decreased temperature, the reaction was not uniform.

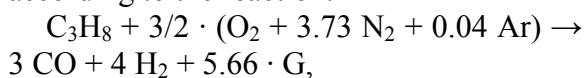
3.2 Gas Analysis

For the gas composition measurements we used GASTEC gas analyzing tubes. Hydrogen cannot be detected directly by

said tubes because remaining hydrocarbons affect readings in big degree.

3.3 Results

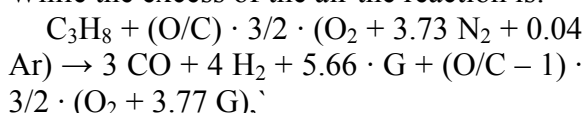
For decomposition of the soy bean oil we used the small reformer (ID=28 mm) with the central core (diameter 20 mm) separated with walls with 4 mm ceramic wool insulation. The first series of measurements was performed with control of CO on the exit. The degree of reformation was estimated as a ratio of measured CO concentration with its maximum level according to the reaction:



where G is a molecule of balance gases – nitrogen and argon, which are not participating in the reaction.

In this case the O/C ratio is unity and the maximum CO concentration is therefore 23.7%.

When the amount of the propane exceeds the stoichiometric ratio some part of the fuel remains not reacted. Without a detailed composition studies we can roughly estimate the upper CO concentration as 24%. While the excess of the air the reaction is:



and it's easy to compute the resulting maximum CO amount:

$$\text{CO (max)} = 3/(3 + 4 + 5.66 + (\text{O}/\text{C} - 1) \cdot 3/2 \cdot 4.77) \cdot 100\%.$$

3. RESULTS AND DISCUSSION

Results of measurements are shown in the Table 1. One can see that a relatively high flow of gas mixture close to a stoichiometric ratio (see Fig.3) brings a higher amount of CO like in tests 3 and 4.

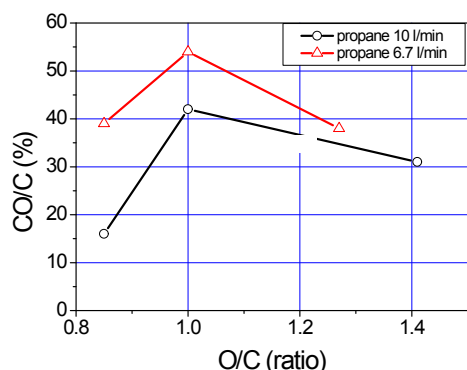


Figure 3: Transformation of C into CO. Propane flow is 10 and 6.7 l/min.

In the same time we controlled the temperature on the external side of the reformation chamber under the outer thermal insulation. This measure doesn't

show the real reforming temperature, but reflected the temperature drift with a time lag. While inside was sometimes over 1000 °C, the readings on the side were only 500-600 °C. This is less than in case of diesel fuel reformation on one-two hundreds degree.

The heat releasing by the partial oxidation of propane is less than of diesel. When the ratio of compounds is close to the stoichiometric the reaction is self-sustaining, its quality depends on heat fluxes and insulation of heat inside the reformer. But effect of the temperature is not direct: a heating of the reformer gives a differently directed trend of CO.

Table 1. Reformation of propane in the self sustaining reaction.

No	Propane, l/m	Air, l/m	O/C	Temperature, °C	CO, vol %	CO _{max} , vol %	CO/CO _{max} , %	CO/C, %
1	3.3	23	0.97	400	8	23.7	34	33
2	5.3	35	0.92	500	8	23.7	34	31
3	6.7	47	1.0	550	13	23.7	55	54
4	10.0	71	1.0	634	10	23.7	42	42
5	10.0	100	1.41	655	6	19.5	31	31
6	10.0	60	0.85	627	4.5	23.7	19	16
7	6.7	60	1.27	605	8	20.8	38	38
8	6.7	40	0.85	590	11	23.7	46	39

Table 2. Reformation of propane with a heating.

No	Propane, l/m	Air, l/m	O/C	Power, W	CO, vol %	CO _{max} , vol %	CO/CO _{max} , %	CO/C, %
9	3.3	18	0.76	196	17	23.7	72	55
10	3.3	20	0.85	210	18	23.7	76	64
11	3.3	20	0.85	168	13	23.7	55	46
12	3.3	22	0.93	210	18	23.7	76	71
13	5.0	30	0.85	140	12.5	23.7	53	45
14	5.0	32.5	0.92	140	16	23.7	68	62
15	5.0	35	1.0	140	13	23.7	55	54
16	6.7	40	0.85	156	10	23.7	42	36
17	6.7	42.5	0.90	156	13	23.7	55	49
18	6.7	47	1.0	156	10	23.7	42	42

The percentage of carbon to CO transferring coincides with CO/CO_{max} when O/C>1, and is equal to CO/CO_{max} · O/C

otherwise, this amount is placed in the last column of the Tables 1 and 2.

Optimal conditions at propane partial

oxidation give 40-50 % of reformation. The observed temperatures were lower than at the processing of diesel fuel. But there we get the reformation degree up to 92 %. The lack of heat can be a reason to the difference of the reaction completeness. We decided to keep the heating current to supply an additional energy to the process. Results are shown on the Table 2.

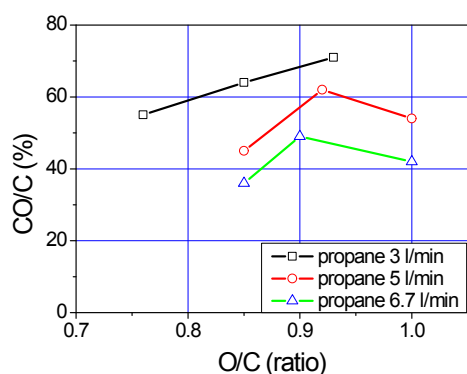


Figure 4: Transformation of C into CO. Propane flow is 3 (196-210 W), 5 (140 W) and 6.7 l/min (156 W).

Graphs for transformation of carbon into CO for the data from Table 1 are given on the Fig.3. One point was removed – it is a test No 11 with the heater power 168 W, it is lower comparing to 210 watts in No 10. Such decrease in power reduces the reformation efficiency on 20 %. For other graphs the power usually was same.

According to the Table 1, without heating the propane flow of 6.7 l/min gives maximum CO yield, next by efficiency was the 10 l/min flow. Using the heating changes the situation. Maximum yield are achieved in mixtures supplied by bigger power of heating per unit of the reforming gas.

The reformation of propane is self sustained in both cases – with and without the additional heating. But the heat released in the reaction is not sufficient for efficient performance. Additional energy input increase the efficiency.

5. CONCLUSION

We designed a new small-scale device for organic fuel reformation. One of its peculiarities is a structure of the active part of the reformer which turns the air and fuel mixture flow manifold increasing the burning front surface and locally decreasing the gas speed in the reformation region. The system doesn't use any catalyst and thus it is not sensitive for fuel impurities. Reformation propane shows up to 54% transferring of carbon into CO without an additional heat supply (using only the heat released in the reaction), and up to 71 % transferring when 210 W are applied. The concentration of CO can reach 18 % in the outgoing gas.

ACKNOWLEDGEMENT

This work was supported by Grant number 2006-C-CC02-P-05-0-000 from the ministry of Commerce, Industry and Energy, Korea. The researchers participating in this study are supported by the grant from “the 2nd phase BK21 project”..

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