Journal of Power Technologies 103 (4) (2023) 243 -- 256

A short review on small steam reformers for methane and other fuels

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Abstract

The article presents an overview of various designs of small reactors intended for steam reforming of methane and other fuels to obtain hydrogen-rich gas. The focus here is mainly on devices with low power and dimensions, intended primarily for laboratory use. Most of the presented steam reformers were used to prepare gases feeding various types of fuel cells. The article also presents research on steam reforming of methane and various alcohols conducted by the Institute of Heat Engineering of the Warsaw University of Technology.

Keywords: steam reforming, steam reformers, steam methane reforming, small steam reformers

Introduction

Steam methane reforming (SMR) is a chemical process in which methane is converted into to hydrogen and carbon monoxide. The steam reforming process takes place in gas-fired steam reformers in which arrange vertically tubes packed with catalyst. The reformer is a main unit in a steam methane reforming system. The operating conditions of the steam reformer are affected by the application and a reformer design. In 1978, Harth et al. [1] proposed a novel system which consists the methane reformer and nuclear reactor so called HTGR (the high temperature gas cooled reactor). In paper [2,3] described a model for methane reformers and compared experimental data with theoretical.

The model proposed by Xu and Froment [3] is an old model but the one more quoted in the literature. This model was used in the simulation of industrial methane reformers [4,5]. In 1982, Murray and Snyder [6] presented a kinetic model of steam methane reformer. This model was a tool to design and analysis a condition system of fuel cells. In 1989, Alatiqi et al. [7] described a heterogeneous one-dimensional model of steam methane reactor in which the heat transfer was by radiation and convection. They did not take into account the formation of coke in the reformer. Tuna et al. [8] studied a biogas steam reforming processes for hydrogen production by system with a fixed-bed reactor (reformer and water-gas shift reactor). Mozdzierz et al. [9] presented the mathematical model of steam methane reformer for solid oxide fuel cell systems. Pulgarin-León et al. [10] developed a model of steam methane reformer based on the model proposed by Xu and Froment [3]. The first steam reformer working at 15 bar pressure came in United Kingdom [11].

A comparative assessment of methods for obtaining blue hydrogen (Fig. 1) using steam methane reforming, autothermal reforming, and natural gas decomposition is presented in [12].

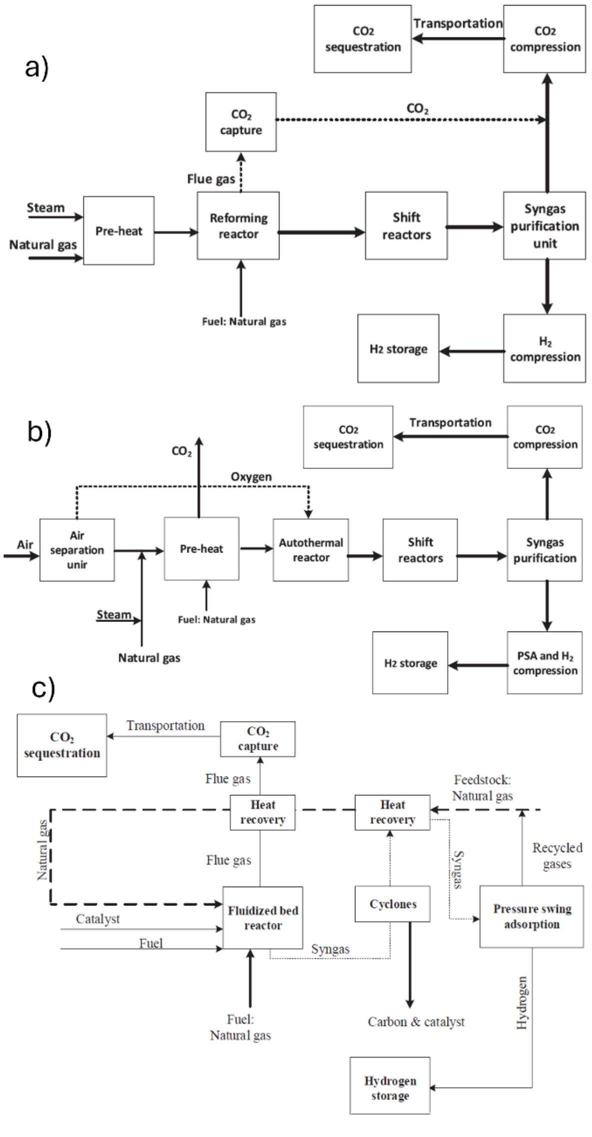

Figure 1: Methods for obtaining blue hydrogen using steam methane reforming (a), autothermal reforming (b), and natural gas decomposition (c) [12]

Steam reforming of methane (and other fuels) to power fuel cells can be carried out in two ways: through external reforming (in reactors called reformers) and through internal reforming directly in fuel cells (here the operating temperature of a given fuel cell must be close to the optimal temperature of reforming process for the fuel used). A study on the performance of SOFC fuel cell fueled by internal reforming of methanol is presented in [13] (see Fig. 2).

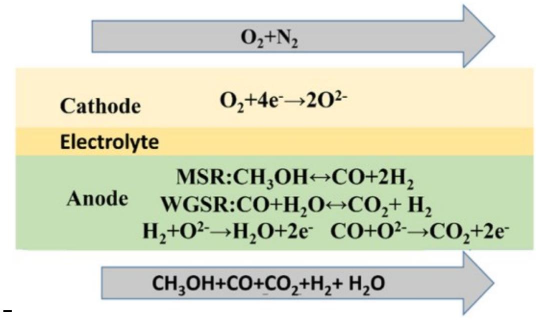

Figure 2: Principle of operation of a methanol-fueled SOFC fuel cell via internal reforming [13]

In [14], an exergy analysis and an environmental impact analysis were carried out for a cogeneration system with a stack of MCFC fuel cells powered by methane in two ways: through internal or external reforming (Fig. 3).

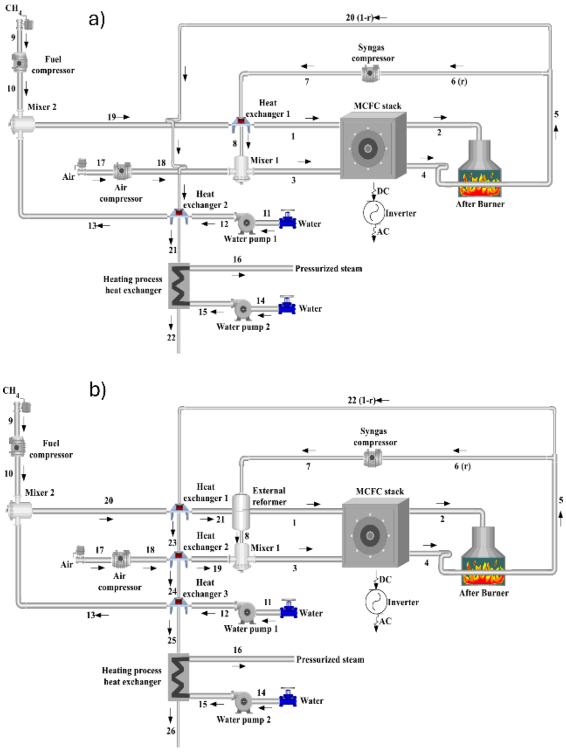

Figure 3: MCFC cogeneration plant with internal (a) and external reforming (b) [14]

An interesting idea for integrating a steam reformer with a gas microturbine was presented in [15]. Thermodynamic analysis showed that such a microturbine can achieve an efficiency of up to 40.7%, which is much higher than the efficiency achieved by gas microturbines of similar power.

A review of previous literature showed that there are many models of steam reformers [7,16–29]. Overview of various mathematical models describing the steam reforming process is summarized and presented in [30].

Review of reformers

The majority of investigations on the SMR process take place in separate reformers in the presence of a catalyst.

Intrinsic rate equations for methane steam reforming with water-gas shift on a Ni/MgA1204 catalyst are reported in [3]. A wide range of precise response mechanisms were examined. Thermodynamic analysis was useful in decreasing the number of plausible causes. Twenty-one sets of three rate equations were kept and used for model discrimination and parameter estimation. The best model has parameter estimates that are both statistically significant and thermodynamically consistent.

[31] employed a dynamic two-dimensional, pseudo-homogeneous dispersion model to predict the performance of a fixed bed catalytic reformer. The link for heat transfer coefficient and effective bed conductivity was shown to have a minimal influence on expected conversions but a considerable effect on wall temperature predictions. There was no definitive assessment of the numerous correlations, and because the wall temperature is critical to the reactor's operating regularity, it was determined that current correlations should be strengthened and certified. The correlations of De Wash and Froment [32] provide the most conservative estimates for wall temperature and were employed in the dynamic simulations. A dynamic model for a steam reformer plant is described [27]. The consequences of a few idealized perturbations are investigated. The model is then used to analyze two sample multi-objective optimization problems for this reactor, yielding Pareto-optimal solutions. A large database of identical optimum control actions under a range of disturbances may be created for subsequent use.

In [33], the reactor was represented by a two-dimensional mathematical model (see Fig. 4) that rigorously accounts for diffusion reaction events inside the particles. Strong radial temperature gradients have been discovered in the reformer tube, especially at the reactor entrance. These temperature variances produce large fluctuations in the methane reaction rate along the radial location, making the catalyst near the reforming tube center underutilized.

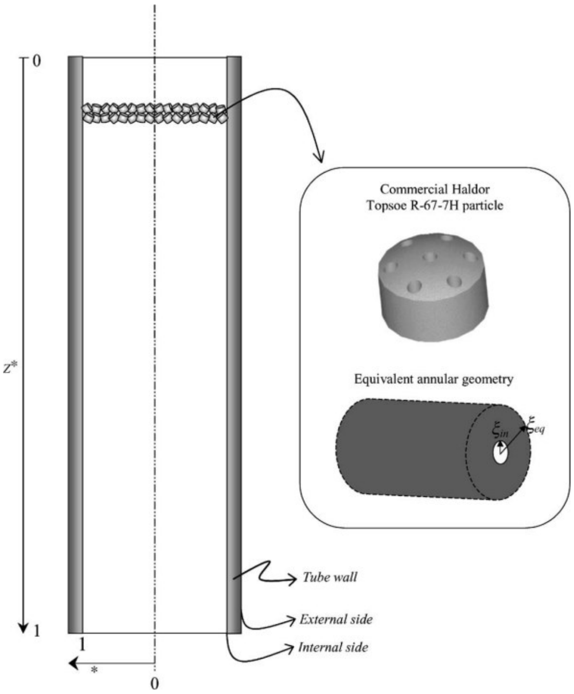

Figure 4: Schematics of the reformer tube, commercial and comparable catalyst particles [33]

A lumped element model of a natural gas steam reformer that is a component of a 5 kW(el.) prosumer fuel cell power plant is shown in [34] (see Fig. 5). The thermal behavior across the whole working range, including plant startup, is described. It is believed that chemical equilibrium exists for the reforming processes, which raises doubts. It is demonstrated how the model was derived, beginning with fundamental thermal and chemical principles. Consequently, model parameters were computed on the presumption of lumped system attributes and have a physical meaning. The parameters were also determined using measured data in order to obtain a better agreement with the trials. To validate the model, experimental data from a demonstration plant was employed.

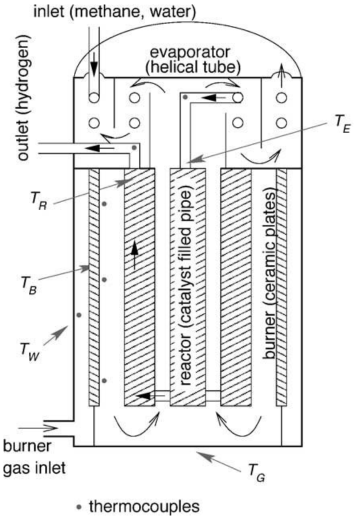

Figure 5: Reformer's cross-section [34]

Using an electrically heated aluminum catalyst (EHAC), a cylinder-shaped methane steam reformer (see Fig. 6) was shown in [35]. Using a differential reactor, an L-H kinetic model for methane steam reforming was presented for Ni/EHAC. Using this kinetic model as a basis, a transient triple-phase lag model was created to look at the processes of internal heat and mass transport. The simulation's output and experimental data from a reactor built like a cylinder contrasted nicely.

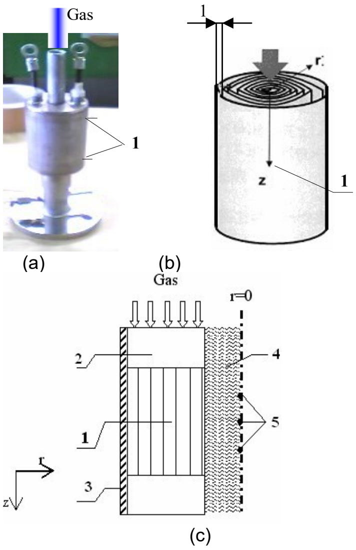

Figure 6: The reactor's schematic (not scaled) [35]: catalyst bed (1), pre-heating section (2), heater (3), thermal insulating material (4), thermocouples (5)

In [36], methane is steam-reformed to power a stack of polymer electrolyte membrane fuel cells (PEMFCs), and this process is optimized for hydrogen production. The auto-thermal membrane reformer consists of two distinct compartments: a methane oxidation catalytic bed and a methane steam reforming bed that houses hydrogen separation membranes (Fig. 7). Drawing from previous practical demonstrations of hydrogen production in such a reactor, a suitable model was built, verified by experimental data, and utilized for optimizing hydrogen generation with respect to reformer efficiency and power output.

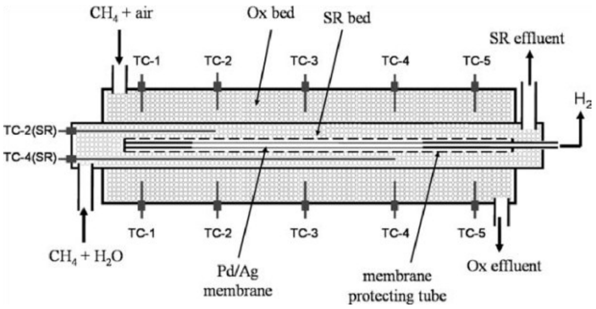

Figure 7: Membrane reformer [36]: Ox – oxidation, SR – steam reforming, TC – thermocouple

In [37], a control-oriented dynamic model of the steam reformer for a SOFC system is described, which incorporates thermal behavior and chemical kinetics. The parameters were found from experimental data using the Breed PSO approach. Simulation findings reveal that the identified dynamic steam reformer model has almost the same performance as the prototype plant. As a result, the model may be used to provide data for the development and testing of a control system in place of the actual steam reformer (Fig. 8). Furthermore, simplified models, such as state-space models and identified models, are available for advanced controller design. The Breed PSO method outperforms the SPSO in terms of global searching capabilities because to the crossover and mutation processes. The suggested identification approach efficiently handles the optimization of operation.

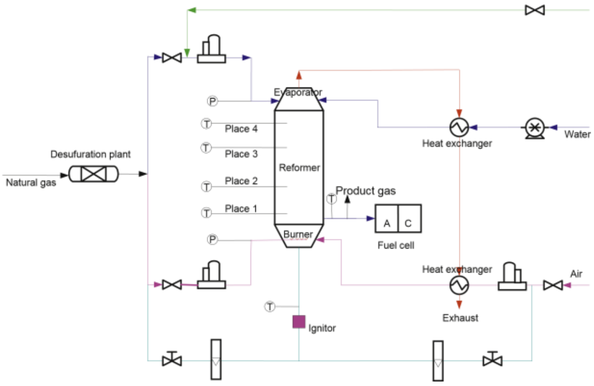

Figure 8: The prototype steam reformer's process diagram [37]

In turn, [38] includes investigations on the kinetics of chemical reactions and a mathematical model of steam reforming of methane and the WGS process in the presence of (18% wt) NiO/α–Al_2O_3 catalyst at 300–700°C and 1 bar pressure. The experiment was conducted out using a plug flow reactor.

[39] provides a computational study of design criteria for a steam reformer using a low-temperature nonreactive heat source. The simulation model was validated using experimental data. To maximize methane conversion rates, heat transfer mechanisms must be tuned for high temperature steam reforming reactions (about 700°C). Additionally, heat source length is a key element to improve.

In paper [40] recommended two types of reformers such as the Pd-Ru membrane reactor [41] and the heat exchanger type reformer [42]. In study [43] was presented a two-dimensional pseudo-homogenous model of the fixed-bed tube reactor for steam methane reforming. In an industrial facility, steam-methane reforming reactions take place in steam methane reformer consist of several hundred fixed-bed reactor tubes filled with supported catalyst particles placed in a high temperature furnace [44–46]. The geometry of the single reforming tube in industrial steam reformers was discussed in [47,48]. The reformer tubes are one of the most expensive plant components in industrial reformers. They are made of metal alloys can creep at high temperatures. It can lead to tube failure and in result expensive tube replacements and production problems. The furnace is used to provide the necessary heat of steam reforming reaction by direct firing (radiant reformers) or by heat exchange with a hot stream (convective reformers). Heat transfer is predominantly by infrared (heat) radiation. The steam reformer is a complex combination of catalyst reactor and heat exchanger. The steam reformer consists with radiant section (furnace) and convection section. Steam reformers are divided on the placement of the burners as top-fired, side-fired and bottom-fired [48,49]. The tubular packed bed reactor characterized a heat transfer coefficient, high steam to methane ratio flow rate and unstable kinetic rate of reaction [50]. Madon et al. [51] used a micro reactor on the improvement of SMR performance in a conventional fixed bed reactor. Furthermore, the steam methane reforming process in the micro reactor depends on the catalyst microstructure, micro reactor geometry configuration, thermal and flow parameters. Literature reported the yield of process steam methane reforming using micro reactor and rare earth catalyst [52–59]. Simpson et al. [60] studied the effect of the reformer operating temperature, pressure, and steam-to-carbon ratio on system efficiency. In paper discusses the single reformer tube model, the furnace model and the computational fluid dynamics (CFD) model [61]. Multiple papers present an analysis and optimization of the methane reforming processes in the reformer reactor including a simulation model [62,63]. Zhang et al. [64] proposed a novel alumite catalyst (electro-thermal catalyst) to steam methane reforming and showed a new kind heat-exchange methane steam reformer. The packed bed reactor was used for the design of steam methane reformer [65]. Khajeh et al. presented a 1-D fluidized-bed tri-reformer reactor in which the tri-reforming process [66]. Franchi et al. [67] has made a comparison the methane conversion ratios in the steam reformer, membrane reactor and reformer and membrane module. Mengi et al. [68] studied kinetics of the steam methane reforming in the 2-D model. Jae et al. [69] optimized the performance of small-scale reformer. Singh and Saraf showed one-dimensional model in axial direction to simulate a work of a side-fired hydrocarbon reformer [70]. Alhabdan et al. [71] presented a heterogeneous model for industrial steam reformers and comprised a number of an side-fired and top-fired industrial reformers. Sinaei Nobandegani et al. [72] presented a one-dimensional model of an industrial steam methane reformer located in Bandar Abbas refinery, using a genetic algorithm. In study [73], showed reformer as a heat exchange plate reactor and studied a simulation on 2D steady-state and dynamic multiphase models for heat and mass transfer. The results of the reformer models demonstrated that plate reformer shows excellent heat transfer performance [74] than a commercial methane steam reformers (a fix-bed reactor) [75].

In [76], three methods of hydrogen production from methane using dielectric barrier discharge plasma-catalysis were compared: direct methane reforming, dry methane reforming and steam methane reforming. Scheme of the research stand for testing these processes is presented in Fig. 9.

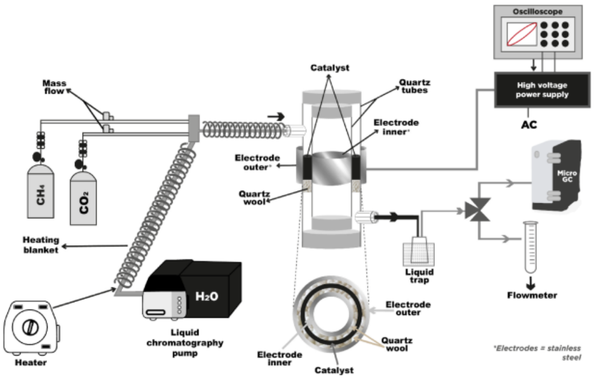

Figure 9: Scheme of the research stand for dielectric barrier discharge plasma-catalysis [76]

The highest methane conversion (24%) and the highest hydrogen content (80%) in the reaction products were obtained for steam methane reforming.

The design of a methanol reformer to power a 3 kW HT-PEMFC (High-Temperature Proton Exchange Membrane Fuel Cell) fuel cell was presented in [77] (see Fig. 10).

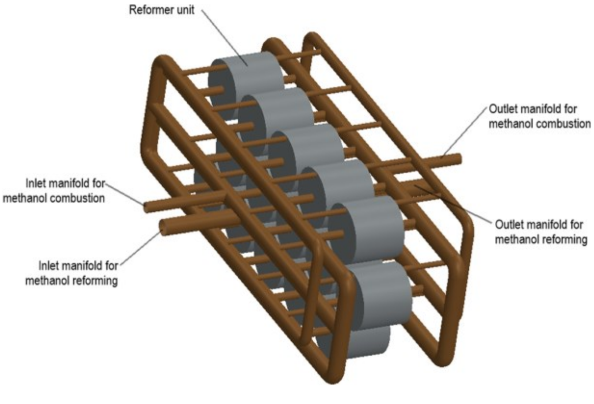

Figure 10: The design of a methanol reformer to power a 3 kW HT-PEMFC fuel cell [77]

In order to select the appropriate operating range of the reformer, CFD (Computational Fluid Dynamics) calculations were performed to predict its performance.

A methanol reformer for powering kW fuel cells was the subject of [78]. By increasing the reformer heating temperature, a methanol conversion rate of up to 99.64% was achieved.

In turn, the thermodynamic analysis of a system with a stack of SOFC fuel cells powered by various fuels (methanol, glycerol, methane and diesel) through an external reformer as a result of the steam reforming or dry reforming process is presented in [79].

Research on steam reforming was also conducted at the Institute of Power Engineering (Poland) and the Institute of Heat Engineering Technology of the Warsaw University of Technology. The Institute of Power Engineering conducted experimental research and the Warsaw University of Technology built a mathematical model, which was published in a joint article [80].

A photo of the reformer with replaceable corrugated catalytic plates, which was the object of the research, is presented in Fig. 11.

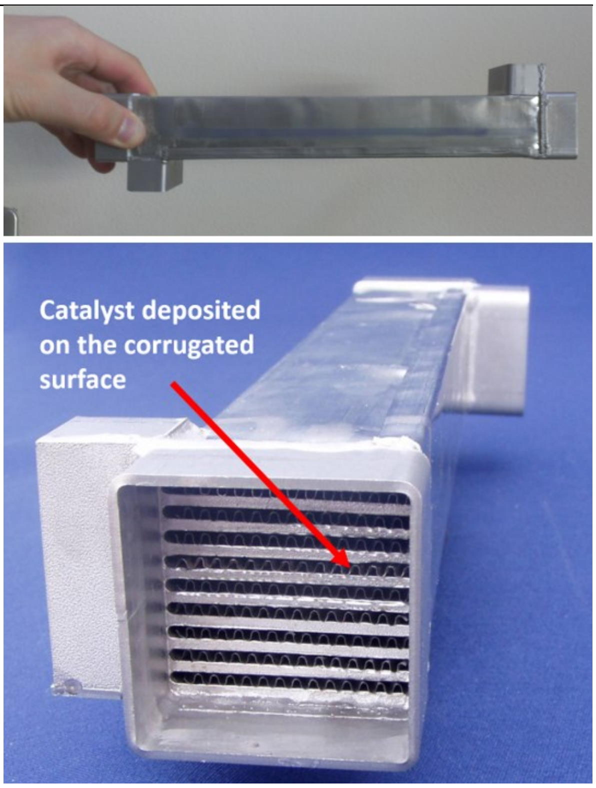

Figure 11: Reformer with replaceable corrugated catalytic plates [80]

There, a model of an external steam reformer used for the production of hydrogen from three different fuels (methane, biogas, and dimethyl ether) was built and validated.

Various studies on steam reforming of methane [81] and alcohol fuels [82,83] were carried out at the Institute of Heat Engineering of the Warsaw University of Technology.

In [81], in order to carry out steam reforming of methane in MCFC fuel cell, a catalyst was placed in the anode channel of the cell - see Fig. 12.

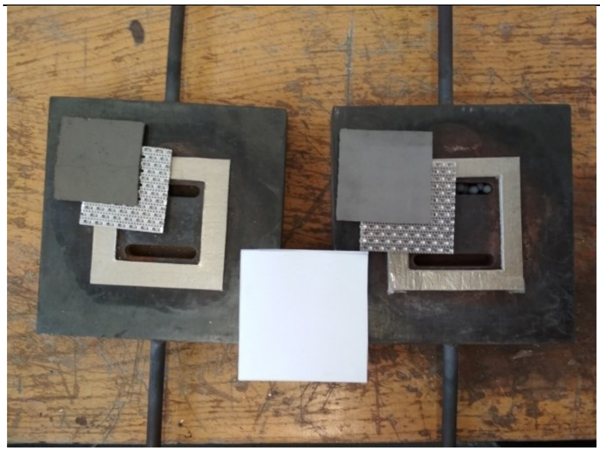

Figure 12: Materials used to build the MCFC fuel cell (catalyst pellets placed at the inlet to the anode channel are visible on the right) [81]

In the case of alcohol fuels [82,83], reforming took place directly on the anode surface of the MCFC fuel cell.

Conclusions

The article presents a comparison of many different types of steam reformers for methane and other fuels. The focus here is mainly on devices with low power and dimensions, intended primarily for laboratory use. Most of the presented steam reformers were used to prepare gases feeding various types of fuel cells. The article also presents research on steam reforming of methane and various alcohols conducted by the Institute of Heat Engineering of the Warsaw University of Technology.

The process of conversion of fuel to hydrogen in steam reformers is mainly influenced by the type of catalyst used, the process temperature, the appropriate and uniform distribution of gases inside the reformer and the residence time of the reactants in the presence of the catalyst.

The temperature of the steam reforming process must be selected appropriately to the fuel (to be converted) and the catalyst used. A very unfavorable phenomenon for the reformer's operation is the deposition of atomic carbon on the surface of the catalyst - the so-called Carbon deposition. This process is harmful because it reduces the active surface of the catalyst, which slows down the reforming process. In order to avoid the phenomenon of carbon deposition on the catalyst surfaces, it is necessary to supply the reformer with an appropriate amount of steam (usually larger than the stoichiometry of the reactions involved in the reforming process).

Acknowledgments

This research was funded by the National Science Center, Poland (Grant number 2020/39/D/ST8/02021).

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