



## Advances in autothermal reformer design



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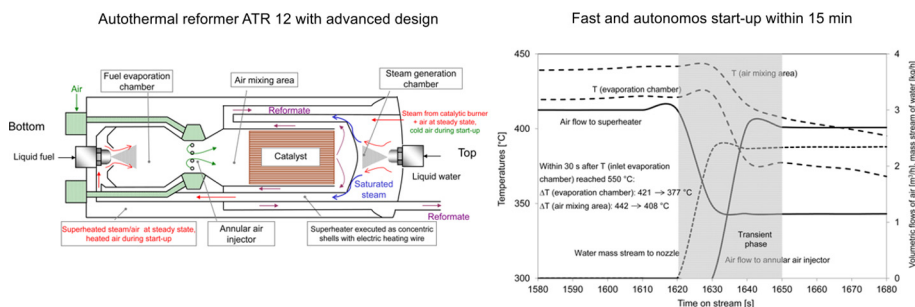
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### HIGHLIGHTS

- Development and experimental evaluation of Jülich's reactor type ATR 12.
- Constructive integration of electric heating wire for fast and autonomous start-up.
- Internal steam superheater modified by concentric shells with low pressure drop.
- New concept for ATR heat management proven to be suitable for fuel cell systems.
- ATR 12 start-up time of approx. 15 min, simplification of fuel cell system layout.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Together with the high-temperature polymer electrolyte fuel cell, the reactor for the autothermal reforming (ATR) of liquid hydrocarbons, such as diesel fuel or kerosene, is the key component of the Jülich fuel cell system in the 5 kW<sub>e</sub> power class. This paper presents some of Jülich's most recent development in the field of ATR reactors, specifically the ATR 12. ATR 12 is characterized by a new concept for the internal generation of superheated steam as one of the ATR reactants using concentric shells instead of coiled tubing and particularly by the integration of an electric heating wire to enable fast and autonomous start-up. Three different experimental procedures for heating up the ATR 12 are presented and discussed, the most suitable of which enables the start-up of the ATR 12 within approximately 15 min. As a consequence, from the system perspective, the bulky start-up burner, which is also difficult to control, along with the corresponding heat exchanger unit, can be dispensed with. Additionally, comprehensive steady-state experiments identify suitable reaction conditions for the operation of the ATR 12.

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## 1. Introduction

Many research groups worldwide have been working on the development of reactor concepts and catalysts for the autothermal reforming (ATR) of liquid hydrocarbon fuels. Active and robust catalysts are essential for the ATR reaction to take place with liquid hydrocarbon fuels. Rhodium, Platinum, Nickel and alloys of Rhodium and Platinum on various supports have often been successfully utilized for this reaction [1–12]. Moreover, many different kinds of liquid hydrocarbon fuels, such as kerosene, conventional and synthetically-produced diesel fuel, as well as various surrogate substances were applied and investigated [13–22]. Some research groups concentrated on using biodiesel, biogas and biomass [23–28]. It was discovered that in comparison to conventional diesel fuels, the ATR of biodiesel fuel requires more oxidative reaction conditions to prevent coke generation. Many papers identified these carbonaceous deposits, along with the adsorption of sulfur-

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containing substances, as the major reason for catalyst deactivation [29–38]. Han et al. [39,40] used an alternate oxidant, hydrogen peroxide  $H_2O_2$ , to supply oxygen and water as reactants for autothermal reforming of diesel fuel for underwater applications (submarines), where there is a lack of oxygen in the environment. They observed corrosion effects in the autothermal reformer materials due to the very high temperatures, when  $H_2O_2$  decomposed in the mixing zone of the reformer. As a consequence, Han et al. incorporated an additional catalytic  $H_2O_2$  decomposer into their reformer design. The same group published a review article about liquid fuel processing technologies and corresponding reactors in the kW-class [41]. Conditioning the mass stream of diesel entering the ATR, i.e., producing a spray of fine diesel droplets, which are then completely vaporized and homogeneously mixed with the other reactants of steam and oxygen, was addressed by a few papers dealing with concepts and technologies for diesel fuel injection [23,42–45]. The success of the concept for fuel injection is regarded as an important condition for the smooth operation of ATR reactors and results in the almost complete conversion of diesel fuel. Ercolino et al. evaluated four different auxiliary power (APU) unit schemes based on a polymer-electrolyte fuel cell stack (PEFC) and ATR and steam reforming of gasoline, light diesel oil and natural gas [46]. They found out that the scheme with steam reforming of natural gas, water-gas shift reaction and methanation of CO was most suitable. Xu et al. present a review about ATR of heavy hydrocarbon fuels on a small scale [47]. They analyzed major problems existing in ATR reactors such as hot spots, formation of coke, and mixing of the reactants. In addition, they present the design of three different ATR reactor types. Pregelj et al. published a model of an APU with PEFC and ATR of diesel fuel, which is based on extensive experimental evaluation of all system components including the ATR reactor [48]. Peters and Samsun present a theoretical study about fuel cell systems with ATR of kerosene for aircraft applications [49]. They analyzed these systems with respect to their multifunctional use in aircraft. Electrical system efficiency, water production and the production of gases for tank inerting were investigated. Also, in our group at Jülich, ATR catalysts and reactors operated with kerosene and diesel have been intensively examined in the past. This research is summarized in several papers and has produced some successive reformer generations [50–54]. Additionally, autothermal reformers were investigated as an important component of an APU for on-board power generation in aircraft or heavy-duty trucks [55,56].

The present paper discusses Jülich's recent reactor generation that is designated the ATR 12. The fast and autonomous heating up of the ATR 12 was investigated by three differing experimental strategies. Furthermore, comprehensive steady-state experiments, which vary some important operational parameters of Jülich's ATR reactors, are presented.

## 2. Reactor development

Fig. 1 depicts the fundamental layout of the ATR reactors recently developed at Jülich. Common to all of these is the fact that in their center there is a ceramic, monolithic substrate (cordierite), which was first coated with  $Al_2O_3$  and  $CeO_2$  to increase the available surface area and then with bimetallic Rhodium/Platinum catalysts to support the ATR reactions. This catalytic system is commercially available. Cold fuel is injected by means of a nozzle into a fuel evaporation chamber. In this chamber, the cold liquid fuel is rapidly heated-up, evaporated and mixed with the other ATR reactants. These are superheated steam (providing the enthalpy flow for heating-up and evaporating the cold fuel) and one part of the volumetric flow of air serving as the carrier gas. During fuel cell system operation, steam comes from the catalytic burner (or from a test rig

device during the stand-alone operation of the ATR). It is conditioned in a superheater, in which the enthalpy flow of the ATR reactions is transferred to the mass stream of steam coming from the catalytic burner. The other part of the total volumetric flow of air is injected into the air mixing area by means of a specifically-designed component termed an annular air injector. The complete mixture of evaporated fuel, steam and air then enters the catalytic reaction zone. A summary of the thermal powers, power densities and specific powers of Jülich's reformer generations prior to the development of ATR 12, the experimental test subject of this paper, is provided in Pasel et al. [51].

The ATR 12 represents a further development of the ATR AH2, whose detailed experimental evaluation was described in Pasel et al. [53]. The most significant difference between ATR 12 and its precursor generations is the way in which the superheater is technically realized (cf. Figs. 2 and 3). The coiled tubing, which in previous reformer generations had the function to transfer the surplus enthalpy flow from the ATR product gas (reformat) produced in the catalytic reaction zone to the saturated steam was exchanged for concentric shells. These concentric shells were additionally equipped with turbulence inserts so as to advance the heat transfer characteristics of the modified construction. From the fluid-dynamic point of view, two important goals were addressed by this: a reduction in the pressure loss in the internal superheater and a more homogeneous mixture of the reactants diesel, steam and air in the fuel evaporation chamber. The new design of the superheater in ATR 12 increases the cross section for the media streams and reduces their path lengths. By increasing the cross section, the flow velocity decreases proportionally and the pressure drop even over-proportionally. In addition, the pressure drop lowers proportionally, when the path lengths of the media streams are reduced. In the design of ATR AH2 (cf. Fig. 3 on the left) saturated steam flows in parallel into three coiled tubings. Depending on the operating status of the reformer, wet steam, i.e., a mixture of saturated steam and liquid water, flows into the coiled tubings. Then, if the reformer is not in a completely vertical position, or if the three coiled tubings are not accurately welded (production tolerances) into the head of the steam generation chamber, liquid water might flow at different quantities into the three coiled tubings. This will lead to different temperatures and densities in the coiled tubings and, as a consequence, to an inhomogeneous mixture in the downstream fuel evaporation chamber. Zones with different steam concentrations will establish. The new design of ATR 12 with its concentric shells (cf. Fig. 3 on the right) overcomes this drawback. Fig. 3, on the right, also shows the turbulence inserts for improved heat exchange and the electric heating wire. Furthermore, from the constructive perspective, the concentric shells provide an adequate space for the installation of an electric heating wire with a thermal power of approximately 2 kW. This additional option facilitates a rapid and autonomous heating-up of ATR 12 and is the most important modification of ATR 12, especially when viewed from the fuel cell angle.

## 3. Experimental

ATR 12 is equipped with a commercial Rhodium/Platinum catalyst.  $Al_2O_3$  and  $CeO_2$  serve as washcoats. Using this catalyst, the ATR 12 can produce a molar hydrogen flow with a thermal power of 28 kW. NEXBTL diesel fuel from Neste (Finland) and SD 10 diesel fuel from Preem (Sweden) were applied as hydrocarbon feedstock. The chemical and physical properties of NEXBTL are described in detail in Pasel et al. [54], while those of SD 10 diesel are given in Meißner et al. [57]. The technical procedure and the apparatus' to analyze the product gas at the outlet of the ATR 12 are described in detail in Pasel et al. [54].

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