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# Development of a high-efficiency hydrogen generator for fuel cells for distributed power generation

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## ABSTRACT

A collaborative effort between Intelligent Energy and Cal Poly Pomona has developed an adsorption enhanced reformer (AER) for hydrogen generation for use in conjunction with fuel cells in small sizes. The AER operates at a lower temperature (about 500 °C) and has a higher hydrogen yield and purity than those in the conventional steam reforming. It employs ceria supported rhodium as the catalyst and potassium-promoted hydrotalcites to remove carbon dioxide from the products. A novel pulsing feed concept is developed for the AER operation to allow a deeper conversion of the feedstock to hydrogen. Continuous production of near fuel-cell grade hydrogen is demonstrated in the AER with four packed beds running alternately. In the best case of methane reforming, the overall conversion to hydrogen is 92% while the carbon dioxide and carbon monoxide concentrations in the production stream are on the ppm level. The ratio of carbon dioxide in the regeneration exhaust to the one in the product stream is on the order of 10<sup>3</sup>.

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## 1. Introduction

As a result of more stringent environmental regulations, hydrogen has attracted much interest as a potential clean energy carrier. Among various methods, steam reforming of hydrocarbons accounts for 95% of the hydrogen produced in the United States [1]. It is expected that steam reforming will play an important role in America's transition to a hydrogen economy.

Conventional hydrogen generation by steam methane reforming (SMR), employing a high temperature reformer (~800 °C), a water gas shift (WGS) reactor (~300 °C) and multiple pressure swing adsorbers, operates at an efficiency around 65–67% based on lower heating value (LHV) of feed and product [2,3]. This conventional SMR system is not very easy to scale down cost-effectively for supplying 35–150 slpm H<sub>2</sub> (to produce 3–13 kW electricity). In recent years,

adsorption enhanced reforming (AER), a novel low-temperature reforming technique, has intrigued significant interest for potential low-cost hydrogen production [4–12]. Thermodynamic analysis conducted by the authors indicates that AER holds the promise of higher hydrogen yield and purity as well as suppressed coke formation [13,14]. Ongoing industrial development effort is being made by Chevron and Intelligent Energy to commercialize this novel technology [15,16]. Hybrid Adsorbent-Membrane Reactors (HAMRs) are also being explored in academia to enhance the process efficiency of hydrogen generation [17].

A collaborative effort between Intelligent Energy and Cal Poly Pomona has recently developed a bench-scale AER-based hydrogen generator for use in conjunction with fuel cells in the small sizes. The hydrogen generator has four beds operated alternately at a lower temperature (~500 °C) than the SMR. It employs potassium-promoted hydrotalcites

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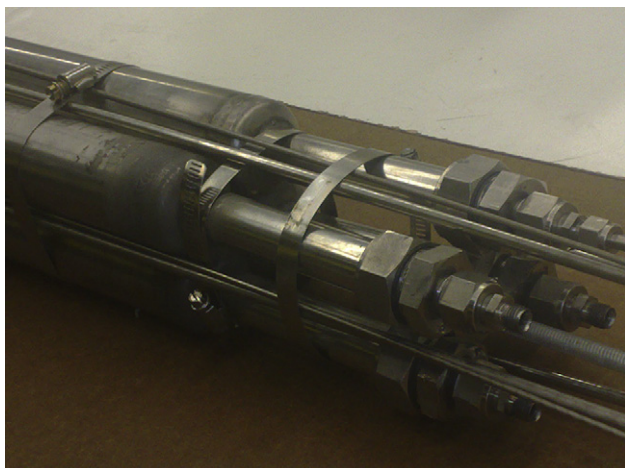


Fig. 1 – Four-tube AER reactors.

(KHTC) as the carbon dioxide capturing material [18–20] and ceria supported rhodium as the reforming catalyst. The removal of  $\text{CO}_2$  from the product favors the reforming and the WGS reactions so that  $\text{H}_2$  is produced in one step with little  $\text{CO}$  and  $\text{CO}_2$  impurities. In addition to steam which is typically used in bed regeneration, this work shows that the  $\text{CO}_2$  purge can also be done using the off gas from the cathode of the fuel cell, which can potentially reduce steam usage. Moreover, it is developed in this work a novel pulsing feed approach to enhance the yield of hydrogen in the production stream. Continuous production of near fuel-cell grade hydrogen has been successfully demonstrated using various feedstocks. In AER of methane, the hydrogen/methane ratio in the product stream is as high as 125 while the  $\text{CO}$  and  $\text{CO}_2$  are on the ppm level. The overall conversion of methane to hydrogen is around 92%.

## 2. Experimental setup

The experimental AER test assembly was constructed with four reaction tubes with a nominal ID of 1.8 inches and a length of 24 inches (see Fig. 1). An external electrical furnace heats the tubes to maintain the proper operating temperatures for the reforming reaction. Solenoid valves under the control of a programmable logic control (PLC) switch the tubes in sequence from a feed of hydrocarbon and steam for the reforming reaction to a purge flow of simulated cathode off gas (COG) or pure steam. At any specific time, one reaction tube would produce hydrogen that flows from the top to the bottom of the bed. The remaining three tubes would have a purge flow in the reverse direction to remove the adsorbed carbon dioxide from the adsorbent. Figs. 2 and 3 show the basic process flowsheet for the experimental unit with the corresponding state diagram for the flow through the tubes. The overall cycle is divided into 24 steps to allow flexibility in controlling the flows through the tubes. For a particular tube the reforming cycle (state P on the diagram) uses six steps (one quarter of the total time) for hydrogen production. Regeneration follows the production cycle in order to remove the adsorbed carbon dioxide. Regeneration takes place in a number of steps. First, a pure steam flows for 10 s (state E on the diagram) in order to sweep excess hydrogen from the tube. A simulated COG then flows (state R on the diagram) for the majority of the regeneration cycle. The oxygen in the simulated COG oxidizes any coke build up. In addition, it shifts the oxidation state of the ceria in the catalyst [21]. A pure steam flow follows the COG (state S on the diagram) to remove any excess oxygen before returning the tube to production. Finally, a backfill flow from the currently producing reactor (state B on the diagram) takes place to minimize the dilution of the hydrogen product when the given reactor resumes hydrogen production.

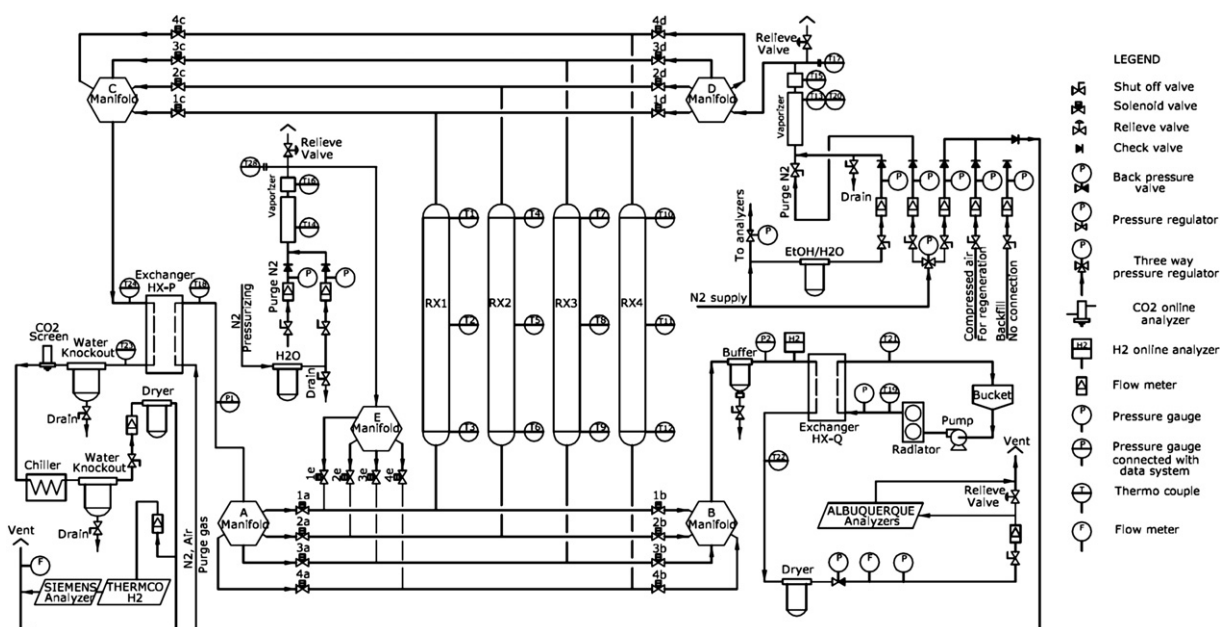


Fig. 2 – A piping and instrumentation diagram of the AER process.

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