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# Experimental investigation on the kinetics of catalytic recombination of hydrogen with oxygen in air

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## ARTICLE INFO

### Article history:

Received 11 April 2014

Received in revised form

27 August 2014

Accepted 31 August 2014

Available online 23 September 2014

### Keywords:

Catalytic recombination

Hydrogen mitigation

Catalytic combustion

Intrinsic kinetics

Palladium catalyst

Recycle

## ABSTRACT

Catalytic recombination of hydrogen with oxygen is one of the most attractive options to control the hydrogen concentration in air. The basic pre-requisite for the process design of any catalytic reactor is the knowledge of kinetic data. In the present study, the kinetic data for the catalytic recombination of hydrogen in presence of 0.5% Pd on alumina catalyst were generated using a packed bed reactor with complete recycle. The experiments were conducted using low concentration of hydrogen in air at different temperatures and the apparent rate constants were estimated assuming a first order reaction with respect to hydrogen. The resistances due to internal and external mass transfer were decoupled from the apparent kinetics and estimated separately. The activation energy and frequency factor were found out using the slope and intercept of the Arrhenius plot. The effect of different process parameters such as temperature, superficial velocity and the catalyst particle size on the overall reaction rate was also studied. The knowledge of the intrinsic kinetics along with the mass transfer can be easily extended for the design of catalytic recombination reactors during scale up.

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## Introduction

Hydrogen and its isotopes have wide applications in the nuclear industry apart from chemical and manufacturing industries [1,2]. Deuterium and tritium will be used as the fuel for International Thermonuclear Experimental Reactor (ITER). Heavy water ( $D_2O$ ) is used as moderator in Pressurized Heavy Water Reactors (PHWR). In most of the nuclear reactors

currently in operation, the coolant used to remove heat from the core is light water or heavy water. Hydrogen is generated in a nuclear reactor core due to reaction of the clad metal (e.g. zirconium) with the coolant water or by the gamma radiolysis of water [3]. The zircaloy clad of the fuel tube in thermal reactors, which contains the fuel pins, is in continuous contact with the coolant water and there is always a chemical interaction of the coolant and clad material. These interactions lead to the production of hydrogen gas and a surface layer of

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<http://dx.doi.org/10.1016/j.ijhydene.2014.08.148>

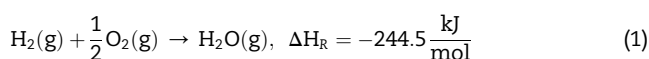
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the metal oxide which adhere to the tube or clad. Such metal water reactions are the primary source for the production of hydrogen inside a nuclear reactor vessel. While the corrosion of the zircaloy with production of hydrogen is a regular phenomenon in the reactor, the problem is of much greater severity in case of a severe accident like coolant failure. The overheating causes fuel and clad temperatures to increase drastically and this in turn leads to enhanced clad-coolant reactions, leading to the production of large amount of hydrogen. The released hydrogen can cause over-pressurization of the containment and potentially an explosion. It can also lead to the formation of an explosive mixture inside the containment and a major fire hazard subsequently [4]. The other source of hydrogen inside the reactor is the radiolysis of water due to the intense radiation field. The interaction of gamma radiation with the coolant and moderator causes its decomposition into molecular hydrogen along with free radical and ionic species [5].

Hydrogen is a very light, mobile gas and its flammability limits in air are 4–76% (v/v) under standard temperature and pressure. In addition, it has very low ignition energy (0.018 mJ) when present with air in optimal proportions [6]. All these factors raise the possibility of a hydrogen induced fire hazard in any facility that handles hydrogen. Severe nuclear accidents have been reported due to hydrogen release, the most recent one being in the Fukushima Daiichi power plant in Japan [7]. In the Three Mile Island accident in USA, partial melt down of the core lead to chemical reactions that caused build-up of a large quantity of hydrogen inside the containment, but there was no explosion because of the absence of oxygen inside the core [8].

There are several methods for hydrogen control in case of any accidental release. The concentration of hydrogen in the atmosphere can be reduced through dilution below the flammability limits by using inert gases such as nitrogen or by increasing the containment volume. Hydrogen can be removed from the atmosphere by purposeful flame ignition at low concentrations or by catalytic recombination with oxygen in air. Hydrogen removal by catalytic recombination is considered in the present study as an effective method, as hydrogen can be oxidized at a temperature much lower than that required for thermal oxidation [3]. Moreover, such reactions can be carried out in passive mode as well especially inside the primary containment of nuclear reactors. In catalytic combustion, emission of nitrogen oxides is completely eliminated as against thermal oxidation and there is no risk of preliminary fire [2].

The recombination reaction is represented by Ref. [3]



In order to design a suitable reactor for the catalytic recombination of hydrogen and oxygen to form water vapour, the thorough understanding of the recombination kinetics is essential. Therefore, a bench scale experimental system consists of a packed bed reactor with complete recycle has been designed and installed. A large set of experiments have been carried out at different operating conditions. The generation of experimental data and the analysis of the results are presented in detail in the subsequent sections.

## Experimental study and analysis

### Procedure of the experiments

A packed bed reactor with complete recycle was used to study the performance of the catalyst. The internal diameter and height of the reactor is 15.76 mm and 300 mm respectively. The catalyst used in the present study was 0.5% Pd (w/w) loaded on spherical alumina particles of 3–5 mm diameter. The fixed bed reactor was filled with 30 g of the catalyst. The process flow diagram of the catalyst test facility is shown in Fig. 1. The other main components in the experimental system were a mixing vessel and a recirculation pump. The reactor was provided with an external heater to carry out the reaction at different temperatures. The temperature of the catalytic bed was monitored at three different locations viz. top, middle and bottom. The average temperature for each run was used for different calculations. The recycle flow rate was adjusted using the bypass valve provided across the recirculation pump and the flow rate was monitored using a rotameter. The operating pressure was measured by bourdon type pressure gauge at the inlet of the reactor. The concentration of hydrogen in the mixing vessel was continuously monitored by a thermal conductivity based gas analyzer. The catalytic reactor was provided with two isolation valves and one bypass valve. During the start-up of a typical experiment, the reactor was isolated initially by closing the inlet and outlet valves. The bypass valve of the reactor was kept open to allow the air to circulate in the loop and the flow rate was adjusted to the required value using the bypass valve of the recirculation pump. Hydrogen was added into the mixing vessel from the cylinder with precise flow control and the concentration of hydrogen was continuously monitored using the hydrogen gas analyzer. The maximum hydrogen concentration in air was limited to 1.5% v/v in the present system. This was mainly to keep the concentration well below the lower explosive limit of hydrogen (4% in air). This also served to limit the heat evolved during the recombination reaction and to maintain nearly isothermal condition of the bed. The packed bed reactor was maintained at a particular temperature by using the heater with a temperature controller. Once the hydrogen concentration was stabilized while the reactor was in isolation, the reactor was taken into the flow path by opening the inlet and outlet valves. The bypass valve to the reactor was

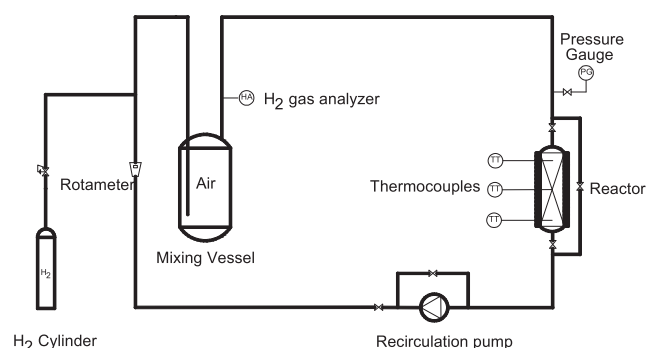


Fig. 1 – Schematic diagram of the catalyst test facility.

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