

Catalytic Combustion of Hydrogen with CO₂/O₂ Using Pt-Coated γ -Al₂O₃ Beads Reactor

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1 Introduction

Recently, due to the energy depletion and the environmental impact, biomass is considered to be a potential renewable energy source for the future [1]. According to the type of desired energy, the biomass can be transferred to various gas, liquid, and solid fuel through chemical or thermochemical process. Among the thermochemical processes, pyrolysis is a promising tool for providing fuel or chemical feedstock. The Pyrolysis is a process of thermochemical conversion in the absence of oxygen at intermediate temperatures, and is a commonly used thermochemical technology for converting biomass into liquids and solids [2]. In commercial pyrolysis process, the pyrolysis gas contains inert, water vapor, and small amount of hydrogen and carbon monoxide is recycled in the reaction chamber. To burn the combustible species in the recycled gas, a combustor is necessary. However, the heat value of the recycled gas is low so the well metered additional fuel and fresh air is added to burn the combustible mixture efficiently. The temperature and oxygen concentration of heated recycle gas need to be well controlled.

The catalytic combustion has been proved to be an essential technique to achieve clean combustion of fuel, meso- and micro scale combustion for portable power sources, and the ultra-lean combustion. The technique promotes oxidation of fuel mixture on suitable catalysts at relatively low temperatures, and thus reducing CO, NO_x and unburned fuel emissions. Hence, the feasibility using catalytic combustion method to burn the combustible mixture in the pyrolysis recycled gas motivates this research. It has been shown that the characteristics of hydrogen reaction on the catalyst surface are completely different from those of the other alkane or carbon monoxide [3][4]. In the first phase, the recycle gas is simulated by artificially blended with hydrogen and carbon dioxide to validate the feasibility of catalytic combustion method.

2 Methodology

To investigate the performance of catalytic combustion method, several conditions are listed in the table 1. The volume concentrations of species in the mixture as well as the equilibrium adiabatic flame temperature are also listed in the table 1. In the case of A-series, the catalyst is verified with various amount of hydrogen oxidized with fresh air stream; in the cases of B-series, the hydrogen is oxidized with stoichiometric oxygen in CO₂ stream. Fig. 1 shows the experimental apparatus and setup. The hydrogen, carbon dioxide and stoichiometric oxygen for hydrogen oxidation are well stirred in the mixing chamber. The temperature is measured by a K-type thermocouple which is mounted on a moving stage. Note that all mixtures are not preheated to check the reaction activity of hydrogen in such kind of mixtures. The fixed reactor is composed of catalytic beads which are prepared using wetness impregnation process with a solution of H₂PtCl₆ to coat platinum on the γ -Al₂O₃ beads. The beads dried and calcined in air at 600°C for 5 hours. The SEM images as well as the EDS analysis are clearly shown the surface and compositions of the catalyst in the Fig. 2.

3 Results and Discussion

The ignition process of hydrogen oxidation in fresh air is shown in Fig. 3. The flow rate of gases is tuned to the case # A-04. Before experiment, the valve 1 is closed; meanwhile, the valve 2 is open for mixture bypass. As the data logger is triggered, the valve 1 and valve 2 are opened and closed at the same time. The thermocouple junction is located at the center point of reactor exit. The result shows that the temperature increases rapidly and reaches a steady value. The increasing rate of temperature of case # B-03 is higher than that of case #A-04. However, despite of the same heat value is input; the final steady value of temperature of case #A-04 is higher than that of case #B-03 due to the different concentration of inert. Basically, the hydrogen mixed with both fresh air and CO₂/O₂ oxidizer can be ignited using platinum catalyst from ambient air temperature without preheating. To further determine the performance of the reactor, the temperature radial profile at reactor exit is shown in Fig. 4. The measured mean temperature distribution shows lower temperature of flue gas near boundary wall of reactor due to heat loss.

The measured temperature at the center point of reactor exit for case A-series and B-series are respectively shown in Fig. 5(a) and 5(b), and the calculated adiabatic reaction temperature is also shown in Fig. 5 for reference. Theoretically, the measured temperature is always lower than theoretical value of temperature. However, as shown in Fig. 5(a), as the concentration of hydrogen is increased in the fuel mixture, the deviation between measured data and theoretical value is also increased for test series A. For the case of B-series, the reaction cannot be sustained as the concentration of hydrogen is lower than 4% in volume. The catalytic reaction is not only affected by concentration of fuel in the mixture but also by residence time. The axial profiles for case #A-04 and #B-03 are compared in Fig. 6. The result shows that the temperature at $x = 5$ mm reaches its maximum value (780.3K) and approximates to adiabatic flame temperature (792.3K). It is believed that the hydrogen is almost consumed and that the hydrogen is oxidized and release heat earlier in the reaction bed within 10 mm. This trends and phenomena are similar to the result of previous research [4]. On the other hand, the temperature distribution of case #B-03 shows contrary trend. The temperature increases as the probe location is increased and approaches a steady value. Note that the deviation between the steady value and the adiabatic flame temperature is 183K.

4 Conclusions

In the present study, the characteristics of catalytic combustion of hydrogen via fixed reactor composed of platinum coated γ -Al₂O₃ beads with fresh air and CO₂/O₂ oxidizer are compared. The axial temperature distribution shows the similar reaction characteristics to the previous investigations for hydrogen oxidation with fresh air. However, the catalytic combustion of hydrogen with CO₂/O₂ mixtures shows totally different axial temperature distribution. Moreover, the deviation between measured data and theoretical estimation for catalytic combustion of hydrogen with CO₂/O₂ mixtures is larger than the case of that with fresh air. Obviously, the clarification of the effect of thermal and

mass transport properties of inert as well as the concentration of oxygen in the mixture on the catalytic combustion are still needed in the future.

Acknowledgement

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Table 1: The experimental conditions

Case no	Flow rate (L/min)			Volume fraction			Eq. T(K)
	Air	H ₂		N ₂	H ₂	O ₂	
A-01	14	0.30		77.34%	2.10%	20.56%	473.73
A-02	14	0.50		76.28%	3.45%	20.28%	584.52
A-03	14	0.70		75.24%	4.76%	20.00%	690.58
A-04	14	0.90		74.23%	6.04%	19.73%	792.30
	CO ₂	H ₂	O ₂	CO ₂	H ₂	O ₂	
B-01	14	0.60	0.30	93.96%	4.03%	2.01%	539.05
B-02	14	0.80	0.40	92.11%	5.26%	2.63%	607.56
B-03	14	0.90	0.45	91.21%	5.86%	2.93%	640.44
B-04	14	1.00	0.50	90.32%	6.45%	3.23%	672.49
B-05	14	1.20	0.60	88.61%	7.59%	3.80%	734.34
B-06	14	1.40	0.70	86.96%	8.70%	4.35%	793.54

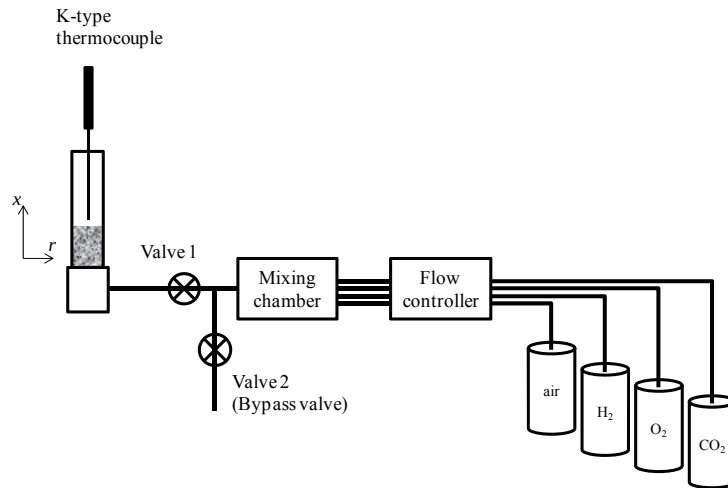


Figure 1. experimental apparatus and setup

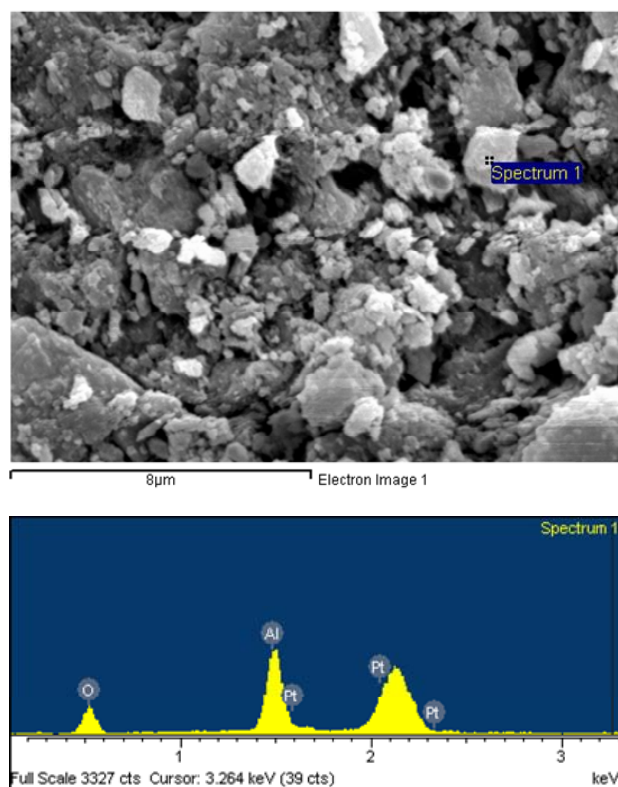


Figure 2. SEM images and EDS spectrum of surface of catalytic beads

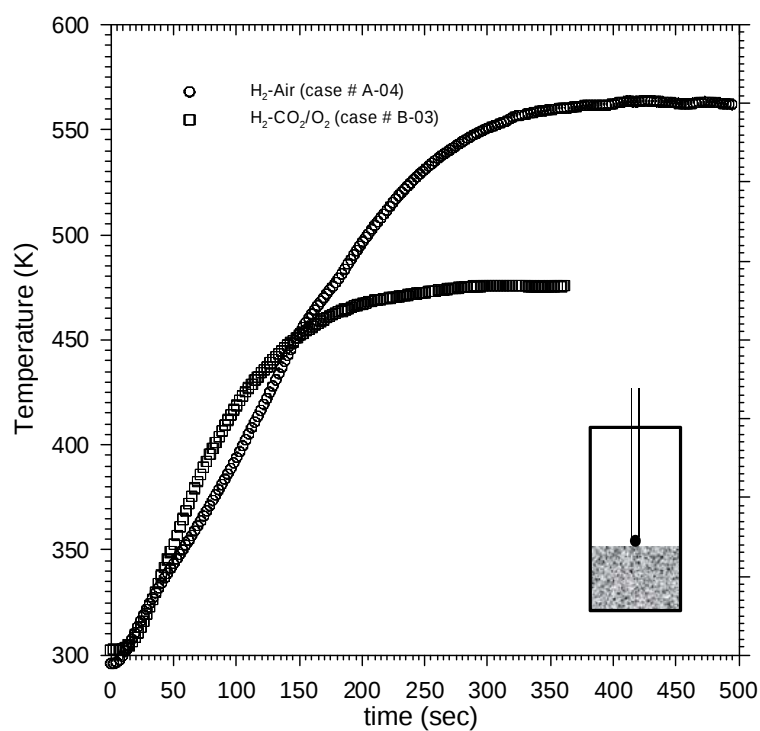


Figure 3.

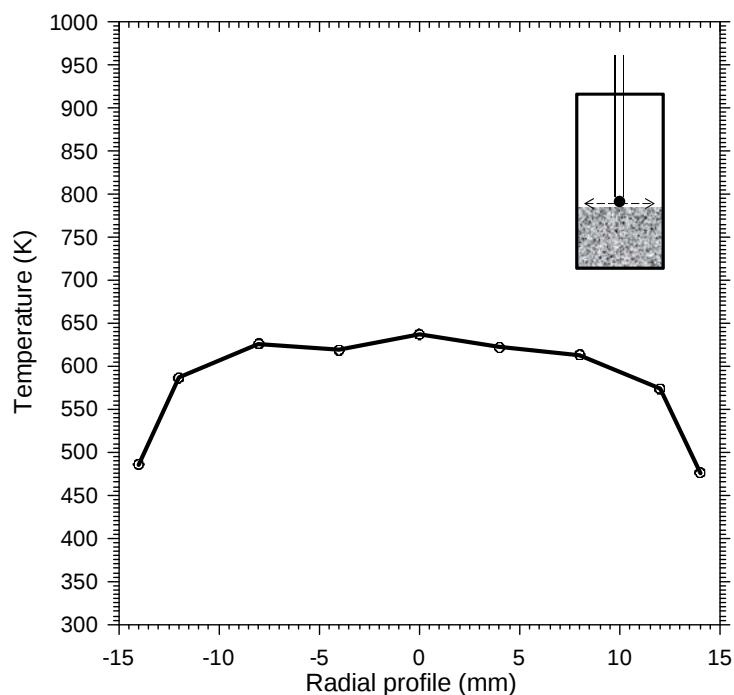


Figure 4. Radial profiles of temperature for case #A-04 at reactor exit

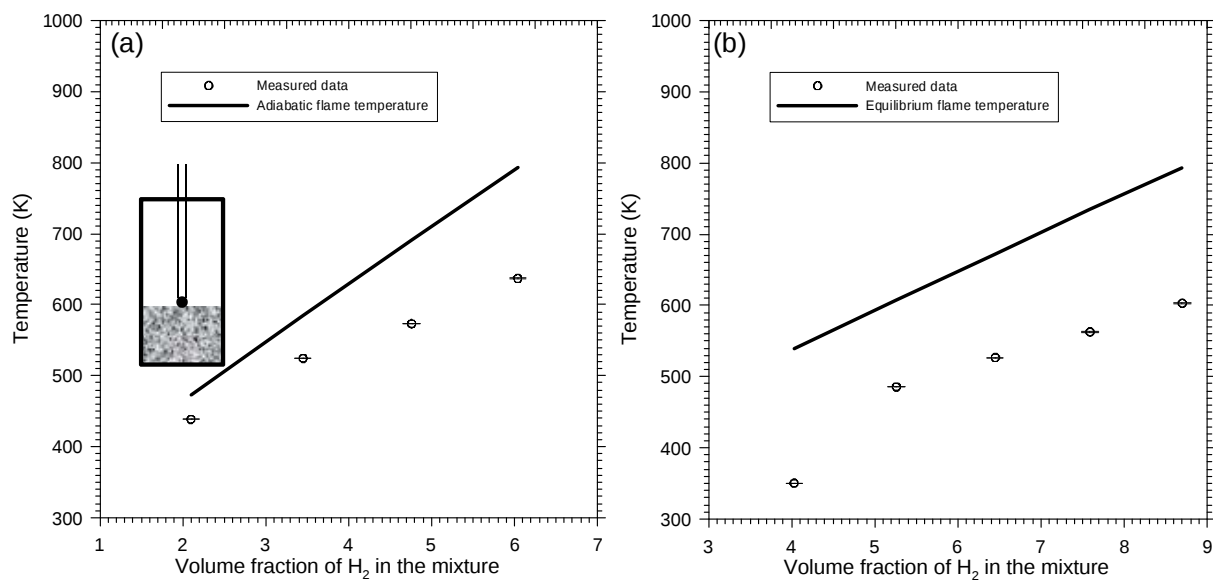


Figure 5. Temperature at the center point at reactor bed exit in function of volume fraction of H₂ in the reactants

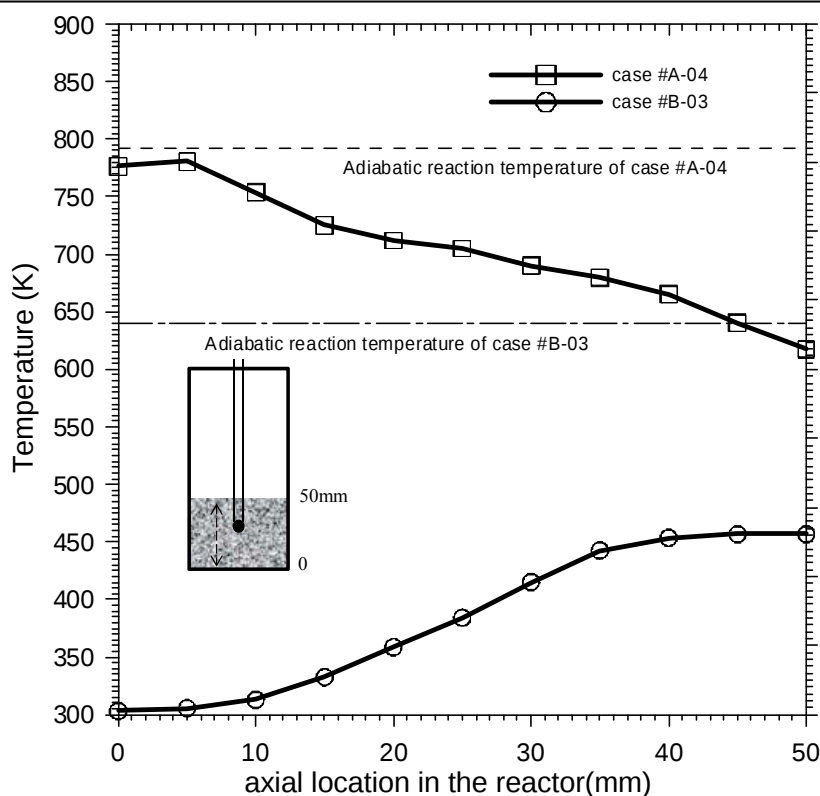


Figure 6. Axial profiles of temperature for case #A-04 and #B-03

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