

Effects of Strain on Transient Ignition Characteristics of Hydrogen-Air Mixtures Impinging on a Platinum Plate

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Abstract

It has long been known that the platinum surface can promote surface reaction and cause combustion of flammable mixtures “without flames”. Recently, catalytic combustion is considered as a successful procedure for improving the stability of lean combustion to minimize NO_x emissions and a technique for enhancing self-sustained combustion to generate low temperature heat in micro-reactors. The characteristic of the ignition process is an important knowledge for improving the design of catalytic combustor or similar systems. Stagnation flows of premixed mixtures impinging on the surface of a heated platinum plate at atmospheric pressure are usually used for studies of the catalytic ignition and combustion. Most researchers discussed the interaction between heterogeneous and homogeneous reactions of hydrogen-air mixture in stagnation flows. Ikead et al. [1] presented four regimes depending on the plate temperature: (1) surface reaction alone; (2) surface reaction inhibiting homogeneous reaction; (3) homogeneous reaction inhibiting surface reaction; and (4) homogeneous reaction alone. Park et al. [2] described that heat generated by surface reactions is primarily responsible for expansion of the flammability limits, and a direct transition to flames occurs over a wide range of composition upon catalytic ignition in the absence of surface heat loss. Recent works have focused on catalytic ignition process in the stagnation flows; various numerical and experimental analyses of the essential surface and gas-phase chemistry have been elucidated and reported [3-6]. However, the role of the strain rate in autothermal catalytic ignition process is still not clear and has not been reported in the literature. This research is to investigate the transient characteristics of the autothermal hydrogen-air mixture catalytic ignition process in platinum stagnation flows with various strain rates. The resultant data will provide useful insight to the design of an improved catalytic combustion system.

Figure 1 shows the schematic experimental arrangement. The platinum catalyst plate consist a foil of dimension 50x50x0.1 mm mounted on a 10x10x3 cm stainless plate. The foil is polycrystalline of purity exceeding 99.99%. To sustain the autothermal condition, isothermal heater is used [4]. Surface and near boundary gas-phase temperatures are measured by K-type thermocouples of 50 μ m in diameter, placed in rear and front of the foil. Steady premixed mixture of hydrogen and air is supplied at a uniform exit velocity through a short

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stainless steel tube. The tube exit is positioned at a distance d below the platinum surface. Numerical simulation is also employed for detailed ignition and flowfield characteristics. The computation is accomplished with the SPIN code [7], which, in turn, uses the TWOPNT [8] software that implements a Newton/Time-Step algorithm. While approaching the ignition point, the Newton iteration is difficult to converge and instead, the transient problem is solved till the steady state is established. For the surface boundary condition, The gas-phase mass flux of each species to the surface is balanced by the creation or depletion of that species by surface reactions. The boundary condition for surface temperature is derived by the balance of diffusive and convective fluxes in the gas phase with thermal radiation and chemical heat release at the surface. The Chemkin software [9] for gas-phase chemical kinetics, and Surface Chemkin [10] for heterogeneous chemical kinetics are used in this study.

Preliminary experimental tests are performed to examine the ignition process under natural heat transfer (without external heating) and isothermal conditions in stagnation flows. Results displayed that two distinct tendencies of the rate of temperature increase exist in these conditions (see Fig. 2). At natural heat transfer conditions, the temperature-increasing curve reveals the trend of one-step continuous to final steady temperature while the combustible mixture is ignited. But at isothermal conditions, results showed the trend of two or even three steps of temperature increase that indicates the further surface or gas phase reaction after ignition. Figure 3 shows the numerical results of catalytic ignition of hydrogen-air mixture. In the process of increasing external power, the surface temperature would increase linearly before ignition and when the power reaches a critical value, the surface temperature rises rapidly to a higher value. Hydrogen would ignite as the surface temperature reached 327K. From Fig. 3 of the surface coverage versus surface temperature, before ignition the main species on the surface is the fuel and increasing surface temperature would lead to a point where the adsorption-desorption equilibrium shifts to desorption, resulting in free surface sites for oxygen adsorption. After ignition, the surface is almost covered by adsorbed species O^* and the system is controlled by mass transport of reactants toward and products away from the surface.

The surface and gas phase temperature at natural heat transfer condition with strain rate $SR = V/d = 40$ 1/s, hydrogen-air ratio range from 7.8% to 20% are measured (see Fig. 4). Results show that gas phase temperature at reaction time $t=450$ sec exceed surface temperature when hydrogen-air ratio above 10% and preheated temperature above 250°C. It reveals that gas phase reactions exist in these conditions. At the same time, reaction temperature is raised with increasing preheated temperature, and the highest temperature can be found around hydrogen-air ratio about 15%. This confirms the results of Bui et al. [5] and suggests that a surface stoichiometric point is observed at about 15% of hydrogen in air. According to above results, hydrogen-air ratio 7.8% and 10% with constant preheated temperature 220°C are examined to investigate the strain rate effects in the catalytic ignition process. For low strain rate ($SR \sim 40$ 1/s) Ikeda et al.'s results [6] indicated the influence on the catalytic ignition temperature is slim. Our research emphasizes on large range of strain rate variation, from $SR=25$ to 150 1/s by varying the distance d and the velocity V . In Fig. 5, from the surface and gas-phase temperature measurements of 10% hydrogen-air mixture, we found that temperature reduces with increasing strain rate, and in the low strain rate conditions (less than $SR=66.7$ 1/s), gas phase temperature is higher than the surface temperature and vice versus for high strain rate cases. The results demonstrate that the strain rate effects on the reaction can be divided into gas-phase dominated and surface dominated regions. At the

conditions of high strain rates (above 75 1/s) and low strain rate (below 53.3 1/s), the gas-phase temperature is almost independent of the velocity. However, at the same strain rate, the gas-phase temperature reduces with increasing separation distance. Figure 6 shows the relationship of temperature-increasing rate and strain rate in the two zones shown in Fig. 2. The results reveal that in the first-step zone (reaction time $t=70$ sec), temperature-increasing rate reduces with strain rate, and the rate for gas phase is higher than for surface. It demonstrates that the surface reaction rises rapidly in a short time, and the surface reaction plays the role of ignition promoter and then gas phase reaction takes over and dominates the rest of the ignition process. Furthermore, at the same separation distance, temperature-increasing rate is approximately a linear function of the strain rate, and at the same strain rate, temperature-increasing rate is reduced with higher velocity. The variation of the surface temperature-increasing rate on the strain rate is indiscernible. However, for the results of the second-step (reaction time $t=450$ sec) zone shown on the right side of Fig. 6, the surface temperature-increasing rate is higher in the low strain rate region ($SR < 66.7$ 1/s). Comparing Fig. 5 and the second-step zone in Fig. 6, for high strain rates, surface temperature is higher than gas phase temperature and the temperature-increasing rate of gas phase is higher than that of the surface. The temperature-increasing rate is reduced with increasing strain rate. In summary, in view of the above results it is believed that, in the low strain-rate region, gas phase reaction dominated the ignition process and the surface temperature is contributed by the gas phase reaction. In the higher strain rate region, the above results show that the temperature-increasing rate of gas phase is slightly higher than surface yet the surface temperature is higher than the gas phase. Higher strain rate of the flow stretches the gas phase reaction layer thinner and pushes the layer closer to the surface that reduces the gas phase temperature and enhances the surface reaction due to inhibited gas phase reaction and enhanced species diffusion to the surface.

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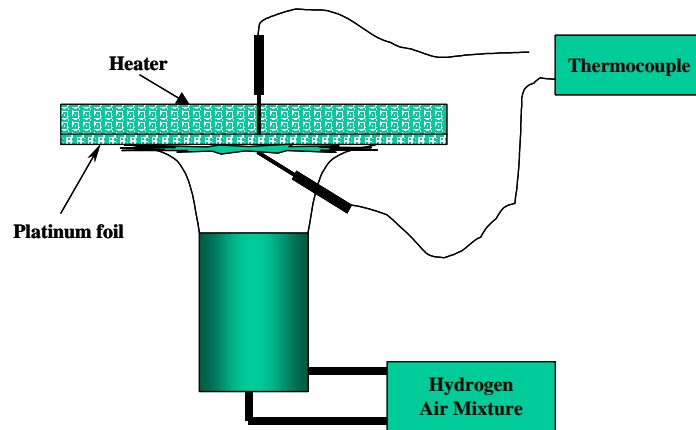


Fig.1 The schematic experimental arrangement

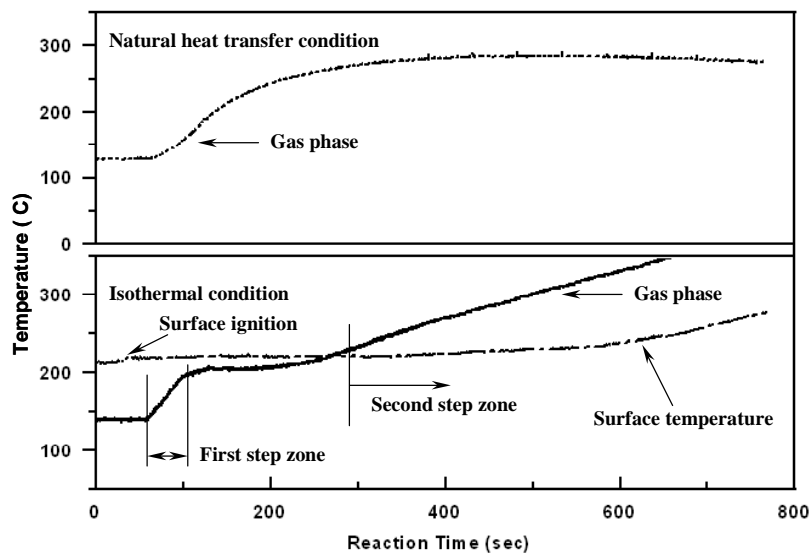


Fig. 2 The relationship between temperature and reaction time in natural heat transfer and isothermal conditions

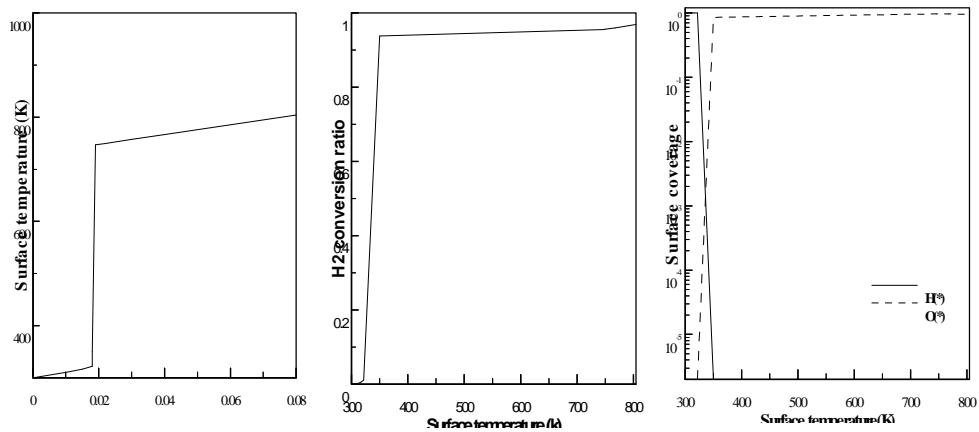


Fig.3 Catalytic Ignition property of H₂ on platinum

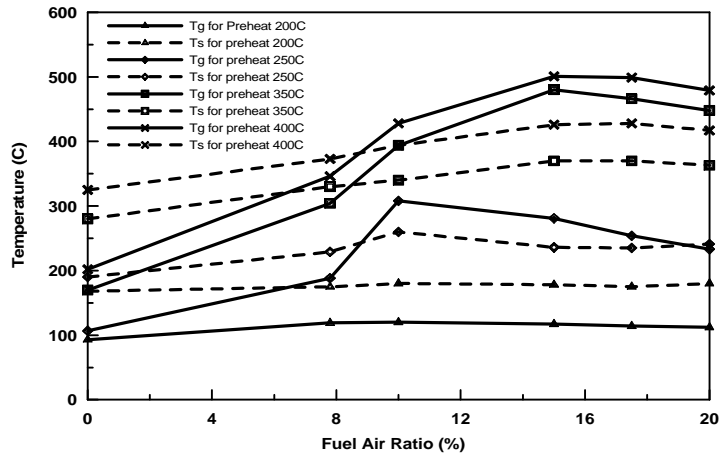


Fig. 4 The relationship between the temperature of surface or gas phase and fuel-air ratio

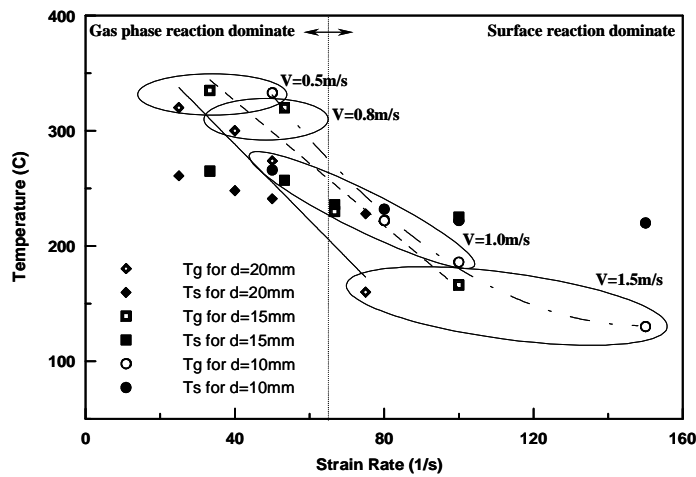


Fig. 5 The relationship between the temperature of surface or gas phase and strain rate

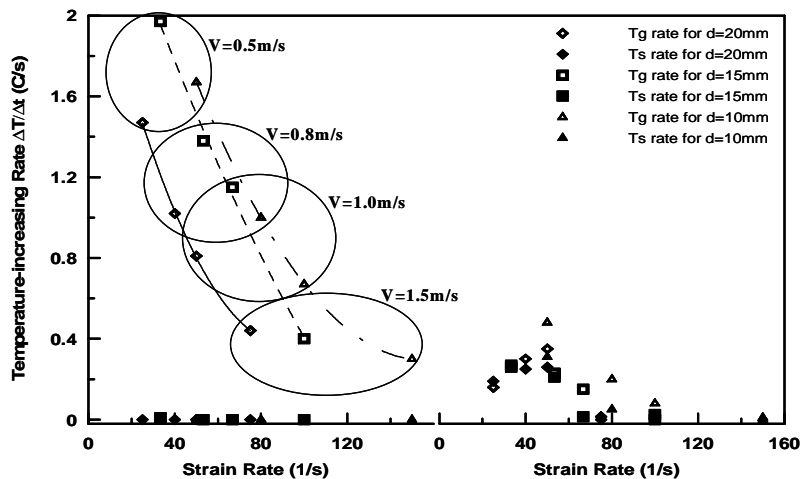


Fig. 6 The relationship between the temperature increasing rate of surface or gas phase and strain rate