# Gradient mechanism on the onset of detonation in the deflagration to detonation transition

Xinmeng Tang<sup>1</sup>, Edyta Dziemińska<sup>1</sup>, A. Koichi Hayashi<sup>1</sup>, Nobuyuki Tsuboi<sup>2</sup> <sup>1</sup>Department of Engineering and Applied Sciences, Faculty of Science and Technology, Sophia University, 7-1 Kioi-cho Chiyoda-ku, Tokyo 102-8554, Japan <sup>2</sup>Department of Mechanical and Control Engineering, Kyushu Institute of Technology, 1-1 Sensui-cho, Tobata-ku, Kitakyushu, Fukuoka 804-8550, Japan

## 1 Introduction

Basal studies have proffered that the main feature leading to the occurrence of DDT is a moderate acceleration of the combustion wave following by an abrupt jump in the flame velocity and a local explosion as the onset of detonation. The onset of detonation occurs locally in the flame zone when the appropriate critical conditions are met. Hot spots that could lead to DDT were analyzed by Merzhanov [1] and later by Zajac and Oppenheim [2]. It was found that although the DDT details may vary, the onset of detonation always seemed to be prompted by an explosion within the explosion. Such a spontaneous-flame hypothesis or gradient mechanism [3] of reactivity suggests that the intrinsic mechanism that triggers a detonation is a spatial gradient of chemical induction time  $\tau$ . If there is a small enough spatial  $\tau$  gradient, a spontaneous reaction wave can propagate fast enough to couple with the compression wave generated by the exothermic reaction and evolve into a self-sustained detonation wave.

In this work, the 2D reactive flow in narrow tubes is numerically solved. An ignition source together with a high pressure zone which furnishes as a shock source are initialized in the narrow tubes filled with hydrogen and oxygen mixture so that a fast DDT can develop based on these conditions. By doing this, two types of autoignition emerge in the preheating zone which are the mild and strong ignitions reported in the experiments. With these autoignitions of different types, the gradient mechanism on the onset of detonation in DDT process is discussed.

## 2 Numerical models and methods

Region	<i>T</i> [K]	p [MPa]	Species
Ignition Source (IS)	2000	2.5	$H_2/O_2$
Shock Relationship (SR)	931	1.699	$H_2/O_2$
Ambient Region (AR)	298.15	0.0739	$H_2/O_2$

Table	1:	Initial	conditions
-------	----	---------	------------

In this work we try to initially place a shock together with a flame in the same direction to speed up the DDT process. The physical model used in this work is set to be similar as that in work of Dziemińska and Hayashi [4]. The tubes are narrow and filled with stoichiometric  $H_2/O_2$  mixture. Initially the tubes are divided into three regions. Among them, ignition source region (IS) is a half-size circle-like region with a high pressure *p* and temperature *T* simulating the flame. Downstream the IS, different *p* and *T* are employed in the shock

Correspondence to: Tang X.M. <a href="mailto:simondonxq@gmail.com">simon@pku.edu.cn</a>, <a href="mailto:simondonxq@gmail.com">shimon@pku.edu.cn</a>

#### Tang, X. M.

relationship region (SR) and the ambient region (AR) in order to offer an artificial shock wave. Table 1 lists the detailed values for these three regions. The values in SR and AR, which decide the strength of the artificial shock wave, are chosen according to the shock wave relation making sure that these settings can allow a direct DDT process within a short distance.

## 3 Results and discussion

## 3.1 boundary layer

With the walls as non-slip and adiabatic, the flow is attached to the walls and gas in the boundary layer is heated more than the interior gas because of the viscous heating. The relationship between the temperature in the interior flow  $T_i$  and on the wall  $T_w$  can be approximated as  $T_w = T_i [1+M_i(\gamma-1)/2]$  where  $M_i$  is the Mach number in the tube flow. Due to the piston effect mentioned above,  $M_i$  is significant, which makes  $T_w$  noticeably higher than  $T_i$ . Figure 1 illustrates the velocity and thermal boundary layers and the corresponding discrete mesh cells in the boundary layers at  $t=3.0 \ \mu$ s. The velocity boundary layer here is about 16  $\mu$ m with a large velocity range from 50 m/s to 780 m/s. The thermal boundary layer, in which the gas is heated by 100 K until now, is thicker than the velocity boundary layer. The mesh cells embedded in the boundary layer are also illustrated in Fig. 1.



Figure 1. Velocity and thermal boundary layers and the discrete grid cells in the boundary layers

## 3.2 First auto ignition in the boundary layer

Pressure and temperature variation near the walls are illustrated in Fig. 2 from 1.0  $\mu$ s to 10.0  $\mu$ s. In the boundary layer *T* has reached almost 1400 K at *t*=5.0  $\mu$ s and over 1400 K at *t*=6.0  $\mu$ s. For H<sub>2</sub>/O<sub>2</sub> mixture the relationship between *T* and  $\tau$  performs approximately an exponential inverse. For instance, with *p* at 1 atm,  $\tau$  is fairly large of over 20  $\mu$ s when *T* is below 1000 K while  $\tau$  is rapidly reduced to about 1  $\mu$ s when *T* rises to 1400 K. It means that at *t*=5.0  $\mu$ s the induction reaction in the boundary layer has started to be at a certain level and at fairly significant rate at *t*=6.0  $\mu$ s. The induction reaction accumulates radicals like H<sub>2</sub>O<sub>2</sub>, H, and O in the boundary layer. Since  $\tau$  is below 1  $\mu$ s, it is expected that within 1  $\mu$ s the heat release process would start. In Fig. 3 one can see the accumulation history of H<sub>2</sub>O<sub>2</sub> species *Y*<sub>H2O2</sub> near the wall. As the piston effect together with the viscosity keeps on heating the gas in boundary layers, H<sub>2</sub>O<sub>2</sub> species accumulates very fast and meanwhile it drifts downstream gradually with the flow. At *t*=7.4~7.5  $\mu$ s, radical concentrations come to critical values and the heat release occurs [5], which indicates auto ignition arise now in the boundary layer. In Fig. 2 one can see that after *t*=8.0  $\mu$ s this autoignition has developed into a significant flame independent of the main flame.



Figure 2. Pressure and temperature variation near the walls from 1.0 µs to 10.0 µs.



Figure 3. Variations of the H<sub>2</sub>O<sub>2</sub> concentration near the wall.

In order to trace how the spontaneous flame develops after the autoignition, Fig. 4 is presented to illustrate the variation of *T* and the concentration of H<sub>2</sub>O<sub>2</sub> species  $Y_{H2O2}$  in five moments from *t*=8.0 µs to *t*=9.5 µs together with the pressure contours. This autoignition is significant enough to be observed from the *T* distribution after *t*=8.0 µs. Ahead of it on the wall there is a region with accumulated high  $Y_{H2O2}$  under the combined effects of *T*, *p*, and flow velocity *V* distribution. According to the gradient reactivity concept, the wave appears when the material spontaneously ignites at the location of the minimum  $\tau$  and spreads by spontaneous ignition at neighboring locations where  $\tau$  is slightly longer. The spontaneous flame develops as a spontaneous reaction wave propagate through the mixture along a spatial gradient valley in chemical induction time  $\tau$ . Here we trace this process by the spatial gradients of *T* and *p* together with the  $Y_{H2O2}$ distribution. The resulting spontaneous wave is analogous to a phase wave and propagates in the direction with the velocity  $D_{sp} = (\partial \tau / \partial x)^{-1}$ . Accumulated radicals such as  $H_2O_2$  species benefit this propagation. In present work, there are significant accumulated radicals in area G1. Such a spontaneous wave creates a pressure wave that later develops into a bow shock, as shown in Figs. 4(b)-(c) and Fig. 2(a) at *t*=9.0 µs. Due to the inversely exponential relationship between *T* and  $\tau$ , for the same variation of *T*, the higher *T* is, the smaller the variation of  $\tau$  would be, which consequently denote a small spatial  $\tau$  gradient. For instance, with

#### Tang, X. M.

#### Gradient mechanism on the onset of detonation in DDT

p=1 atm,  $\tau$  at T=1300K, 1400 K and 1500 K are 2.03 µs, 0.68 µs and 0.38 µs, respectively. With the same variation of *T*, the variation of  $\tau$ , namely the  $\tau$  gradient, are much smaller under higher *T*. From Fig. 2 one can see the *T* gradients at t=8.0 µs and 9.0 µs. The *T* in front of the spontaneous flame varies in the range of 1200~1400 K at t=8.0 µs. Consequently, significant fluctuations of *T* at relatively low temperature range result in significant spatial  $\tau$  gradients. The spatial  $\tau$  gradients at t=9.0 µs are worse. In areas G2~G3, the amount of accumulated radicals is also very limited. As a result, this spontaneous reaction wave captures a  $D_{sp}$  which is much smaller than the speed of the bow shock due to large spatial  $\tau$  gradients and a lack of accumulated radicals. The coupling of traveling shock wave with the chemical energy release cannot be realized. The bow shock propagates far away from the spontaneous reaction wave and merges with the precursor shock, as shown in Fig. 4(d).



Figure 4. *T*, *p* and  $Y_{H2O2}$  at five moments from *t*=8.0 µs to *t*=9.5 µs. For each moment *T* and  $Y_{H2O2}$  are axisymmetrically shown in one frame. G1~G3 mark the effective area of *T* and  $Y_{H2O2}$ . Black lines indicate pressure contours.

## 3.3 Second autoignition as a hot spot and the onset of detonation

The first autoignition near the wall does not lead to a onset of detonation due to the large spatial  $\tau$  gradients and corresponding small speed of spontaneous reaction wave. Nevertheless, it generates pressure waves and benefits the preheating strength of the precursor shock. Also, now the area of flame fronts is greatly enlarged. The flame acceleration and heat release rate are substantially promoted. In addition, the products capture a velocity opposite to the wall, which narrows the effective diameter of the tube. This allows the compression effect, namely the piston effect, to be more focused on the reactive mixture between the flame fronts and the precursor shock. Due to these issues, gradually in this area *T* and *p* are built up. Figure 5 demonstrates the pressure and temperature variation on the tube axis from 10.6 µs to 11.1 µs. The mixture temperature behind the precursor shock reaches 1300~1400 K with a flat gradient and *p* reaches over 2.0 MPa. An autoignition, as shown in Figs. 5(b) at *t*=10.7 µs to 10.8 µs, arises in this region with the same mechanism as discussed above which is the radials accumulate and the heat release proceeds. The emergence of this autoignition can be manifested as well in Fig. 6 marked as *A*.

Compared with the first autoignition in the boundary layer in section 3.2, present autoignition is provided with higher *T* and *p*. Moreover, around this spontaneous reaction point *A* the spatial  $\tau$  gradient is very small in the vertical direction. As shown in Fig. 6 at *t*=10.9 µs, around *A* exists a wide valley with small spatial  $\tau$  gradient, marked as *B*. The pike *T* of *A* is 1670 K while *T* in *B* is 1400 K. the corresponding *p* are about 1.8 MPa and 1.7 MPa. As mentioned above, due to the inversely exponential relationship between *T* and  $\tau$ , for the same variation of *T*, the higher *T* is, the smaller the variation of  $\tau$  would be, which consequently denote

a small spatial  $\tau$  gradient. Pressure is also very important in this process. High pressure helps largely reduce the  $\tau$  and correspondingly the spatial  $\tau$  gradients can be much smaller. Present high pressure over 1.7 MPa further decrease the spatial  $\tau$  gradient in this gradient valley *B*.



Fig. 5. Pressure and temperature variation on the tube axis from 10.6 µs to 11.1 µs.



Fig. 6. Temperature profiles in the preheating zone from 10.7  $\mu$ s to 11.1  $\mu$ s together with the temperature contours. The hot spot is marked by the yellow circle as *A*. Two black arrows indicate the the path of minimum temperature gradients around the hot spot in the spatial  $\tau$  gradient valley *B*.

According to the gradient mechanism, the spontaneous reaction wave propagate through the reactive mixture along the spatial  $\tau$  gradient direction with the velocity  $D_{sp} = (\partial \tau / \partial x)^{-1}$ . In this work the minimum spatial  $\tau$  gradient is located in valley *B* and in this direction the spontaneous reaction wave capture the maximum speed. This is exactly what is manifested in Fig. 6. One can see that reaction wave originated from *A* propagates in all directions while along valley *B* its speed is the fastest.

In Fig. 7 more details are surfaced by the history of density gradient  $\nabla \rho$ . The spontaneous reaction wave spreads and produces expanded products which results in a surrounding shock wave. In the vertical direction, the spontaneous reaction wave spreads much faster than the shock wave, which implies that in valley *B* the spatial  $\tau$  gradient is small enough to make  $D_{sp}$  fall in the interval roughly limited by the speed of sound and the Chapman-Jouguet detonation velocity  $D_{CJ}$ . When this critical spatial  $\tau$  gradients are achieved, the reaction wave can catch up with the shock wave, which happens at *t*=11.08 µs as shown in Fig. 7(e). The coherent coupling of the traveling shock wave with the chemical energy release is realized. And the coupled

#### Tang, X. M.

wave can be called detonation which begins to emerge after  $11.08 \ \mu$ s. With this local detonation, the DDT would be achieved shortly, as shown in Fig. 7(i).

With a gradient valley *B* where the spatial  $\tau$  gradient is small enough, autoignition *A* eventually accomplish the onset of detonation and subsequent DDT in the narrow tube. We can call *A* as an active hot spot which is assembled with high *T*, *p* and a small spatial  $\tau$  gradient in some directions. In the formation of an active hot spot, how these critical conditions are build up is a significant question which requires special discussion.



Fig. 7. Onset of detonation from 10.7  $\mu$ s to 11.1  $\mu$ s demonstrated by  $\nabla \rho$ 

## 4 Conclusions

Two different types of autoignition are simulated and analyzed in this work: the first one occurs in the boundary layer due to continuous wall heating and the other one in the preheating zone. The gradient mechanism of reactivity is substantiated. And the critical condition that an active hot spot requires is surfaced which is high pressure, high temperature and also a small enough spatial  $\tau$  gradient in a certain direction around it.

## References

[1] Merzhanov AG. (1966). On critical conditions for thermal explosion of a hot spot. Combust. Flame. 10(4): 341-348.

[2] Zajac LJ, Oppenheim AK. (1971). Dynamics of an Explosive Reaction Center. AIAA J. 9(4): 545-553.
[3] Y.B. Zeldovich. (1980). Regime classification of an exothermic reaction with non uniform initial conditions. Combust. Flame. 39: 211-214.

[4] Dziemińska E, Hayashi AK. (2013). Auto-ignition and DDT driven by shock wave-Boundary layer interaction in oxyhydrogen mixture. Int. J. hydrogen energy. 38(10): 4185-4193.

[5] Liu F, Guo H, Smallwood GJ, Gülder ÖL. (2002). Numerical Study of the Super-Adiabatic Flame Temperature Phenomenon in Hydrocarbon Premixed Flames. Proc. Combust. Inst. 29(2): 1543-1550.