Research paper

Study on laser ignition of boron / potassium nitrate in vacuum

Hiroyuki Koizumi*†, Masakatsu Nakano**, Takayoshi Inoue*, Masashi Watanabe***, Kimiya Komurasaki****, and Yoshihiro Arakawa*

*University of Tokyo, Department of Aeronautics and Astronautics, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, JAPAN
† Corresponding address: koizumi@al.t.u-tokyo.ac.jp

**Tokyo Metropolitan College of Aeronautical Engineering, 8-17-1 Minami Senju, Arakawa-ku, Tokyo 116-8523, JAPAN

***Nichiyu Giken Kogyo Co., LTD., 21-2 Matoba-shinmachi, Kawagoe, Saitama 350-1107, JAPAN

****University of Tokyo, Department of Advanced Energy, 5-1-5 Kashiwa-no-Ha, Kashiwa, Chiba 277-0882, JAPAN

Received: July 27, 2006 Accepted: September 15, 2006

Abstract
Laser ignition characteristics of boron / potassium nitrate in vacuum was investigated using a diode laser as an energy source. Ignition probability in vacuum was experimentally measured and the minimum laser power for the ignition was at most 0.4 W. The result was compared with the numerical calculation, three dimensional heat conduction inside the pyrotechnic with the exothermic reaction. As a result, the calculated threshold power and ignition time well agreed with the experimental results.

Keywords: Laser ignition, Vacuum, Boron, Potassium nitrate, Diode laser

1. Introduction
Boron is frequently used as a fuel of rocket motors and delay composition of pyrotechnic products. Mixture of boron with potassium nitrate (B/KNO_3) has been widely used as an ignition systems of rocket motors and airbags in vehicles. In recent years, it has been used for the propellant of micro space propulsion system [1)-4)], and it is strongly required to clarify the ignition characteristics of B/KNO_3 in vacuum.

Ignition of pyrotechnics by means of laser beam is one of the promising methods to initiate combustion of energetic materials. The laser ignition benefits from the high energy intensity, electrical insulation, precise control of the heating time, and direct transfer of the energy into pyrotechnics. Diode lasers have great advantages among a number of lasers due to their compactness, cheapness, and high energy conversion efficiency.

Several researchers have studied on the laser ignition of pyrotechnics [5)-11)]. They observed that the ignition depends on a thermal mechanism, and the incident laser energy and the laser power density control the ignition threshold. They also noticed that the laser energy required for the ignition decreases as the laser power is increased. However, most of them have conducted the experiments in the atmosphere. Only a few studies were carried out in the lower background pressure [7)].

In this study, we clarify the ignition characteristics of B/KNO_3 by diode laser in vacuum, which is essential for the application to the fuel of spacecraft rocket. In addition, experiments in vacuum will reveal the nature of the ignition mechanism by removing the effect of atmosphere.

2. Reaction processes of B/KNO_3
Reaction processes of boron and potassium nitrate had been well investigated by Yano in 1980s [12), 13)]. He concluded that the significant heat is produced in the reaction process

$$B + KNO_3 \rightarrow KBO_2 + NO \qquad (1)$$

It is the reaction of condensed phase potassium nitrate and boron particle. Boron has the melting point of 2340 K and potassium nitrate has the melting point at 612 K. Yano showed by his thermochemical analysis that the above exothermic reaction occurred between 720 and 810 K[12]. There boron particles are oxidized with liquefied potassium nitrate around. The oxidation forms the oxide shell around the particle surface, which prevents the oxidation. The reaction of boron particles is terminated when the thickness of the oxide shell reaches to certain value. The above exothermic reaction is not largely effected by background pressure because the main reaction is not gaseous phase reaction but condensed phase reaction. Yano showed that the burning rate of B/KNO_3 with high boron weight fraction is insensitive to the background pressure, namely the pressure exponent is close to zero.

3. Experimental setup

3.1 Boron / potassium nitrate

Boron / potassium nitrate used in this study was supplied from Nichiyu Giken Kogyo Co., Ltd.. Averaged diameter of the mixed boron particle was less than 1.0 μm. The weight fraction of the boron particles is 0.30. The mixture is compressed in cylindrical shapes with the diameter of 3.2 mm and the height of 2.0 mm.

3.2 Diode laser optical system

Figure 1 shows the schematic diagram of the experimental setup. Diode lasers used in this study have the maximum beam power of one Watt and the wavelength of 808 nm. Focusing assembly consisted of a 10-mm-diam lens pair and 15.7-mm-diam focusing tube optics. The focal length can be adjusted by changing the position of the lens pair, and here the typical length was 15 to 16 mm.

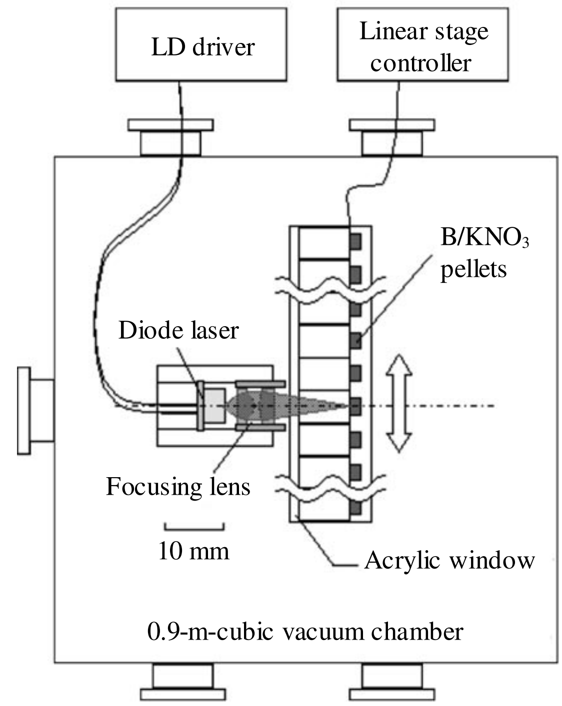

Fig. 1 Schematic diagram of the experimental setups.

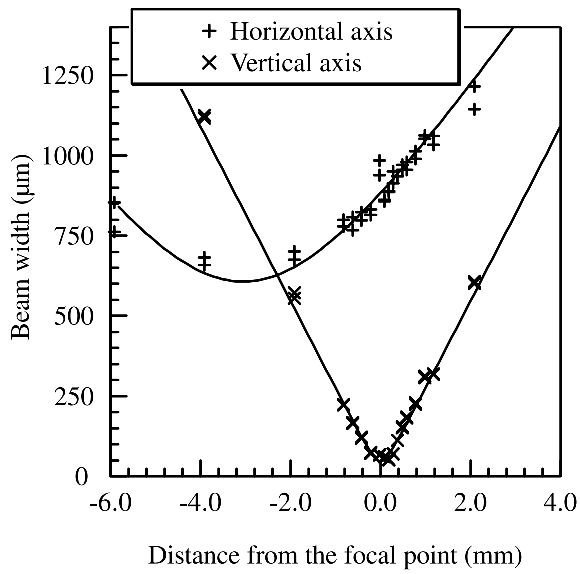

Fig. 2 Profiles of beam widths along the laser beam axis measured by a beam profiler.

Beam profile from the diode laser was measured using a 2D CCD beam profiler. Beam profile of diode laser can be roughly assumed as Gaussian beam with different beam widths on vertical and horizontal axes. Those widths were determined from the measured beam profile. Figure 2 shows the change of beam widths along the laser beam axis, where beam width means the e^{-2} power point in the profile. The laser beam intensity had the maximum of 740 W cm^{-2} and the FWHM of 0.59 mm.

3.3 Pyrotechnics feeding device

The effect of laser power on the ignition probability was investigated. Figure 3 shows the picture of the feeding system of pyrotechnics for laser beam. Twenty pellets are installed into holes aligned on a line, and each pellets are separated by plastic (polyoxymethylene) walls. Laser beam enters through an acrylic window which prevents the optical system from the fouling. The stage on which the pellets were installed was displaced after every shot using a linear transverse stage. The distance from the diode laser to the surface of the pyrotechnics was set to give the strongest irradiation on the surface. The variation of the position of the pellets was at most ±0.2 mm (standard

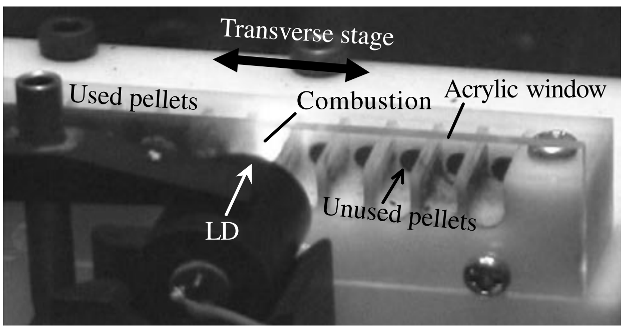

Fig. 3 Picture of the experimental setups.

deviation), which was measured using a laser displacement sensor.

All experiments in vacuum were performed using a 0.9-m-cubic vacuum chamber. The chamber is made of stainless steel and evacuated using two stage pumps: rotary pump and turbo molecular pump. The base pressures were between 1–2 × 10^{-3} Torr.

4. Numerical calculation

4.1 Calculation method

Three dimensional heat conduction problem with the exothermic reaction as heat source was numerically solved to confirm the ignition mechanism. A rectangular solid is selected as the calculation domain. Incident power by laser beam was treated as heat flux into the boundary surface. Exothermic reaction was included as assuming that the reaction rate is constant and it starts when the point reaches to the reaction temperature. Of course, assuming Arrhenius type chemical reaction would be better. However, the parameters appeared in Arrhenius equation of the reaction (2) was no available. In stead, the reaction rate of the reaction was estimated using the measured burning rate in the elsewhere [16]. The estimated rate was 90 m s^{-1}, at most 130 m s^{-1}. The calculation was conducted with changing that rate.

The governing equation is

$$\rho c_p \frac{\partial T}{\partial t} = k\nabla^2 T + \rho \Delta h_R \omega\, s(t,x)$$
$$\text{for } 0 \le x \le L_x,\ 0 \le y \le L_y,\ 0 \le z \le L_z \qquad (2)$$
$$\text{B.C.}\quad \left.\frac{\partial T}{\partial z}\right|_{z=0} = -\frac{\alpha\, q(x,y)}{k} \quad , \quad \left.\frac{\partial T}{\partial n}\right|_{\text{other boundary}} = 0$$

where x is taken along the horizontal axis and y is along the vertical axis, and z is along the beam axis (see Fig. 4). Second term on right hand side of equation (2) expresses the exothermic reaction, where Δh_R is the heat of reaction, ω is the reaction rate (constant), and $s(t, x)$ is a function which determines the reaction period, defined as.

$$s(t,x) = \begin{cases} 1 & , \quad t_R < t \le t_R + \omega^{-1} \\ 0 & , \quad \text{otherwise} \end{cases} \qquad (3)$$
$$t_R\text{: the time when } T(t,x) \text{ reaches to } T_R$$

It means that the reaction at a certain point, x, starts when the temperature $T(t, x)$ reaches to T_R and it lasts during ω^{-1}. The calculation domain is taken as a quarter of the rectangular solid from the symmetry. Laser beam irradiates $z = 0$ plane with the intensity profile

$$q(x,y) = Q\frac{4}{\pi w_x w_y}\exp\left\{-\left(\frac{2x}{w_x}\right)^2 - -\left(\frac{2y}{w_y}\right)^2\right\} \qquad (4)$$

where Q is the incident laser power, w_x is the horizontal beam width and w_y is the vertical beam width at the focal point. The laser energy was absorbed with the conversion efficiency α, where we assumed all of the energy was absorbed, namely $\alpha = 1$.

Figure 4 shows the calculation domain and grid with irregular mesh. Space discretization was performed

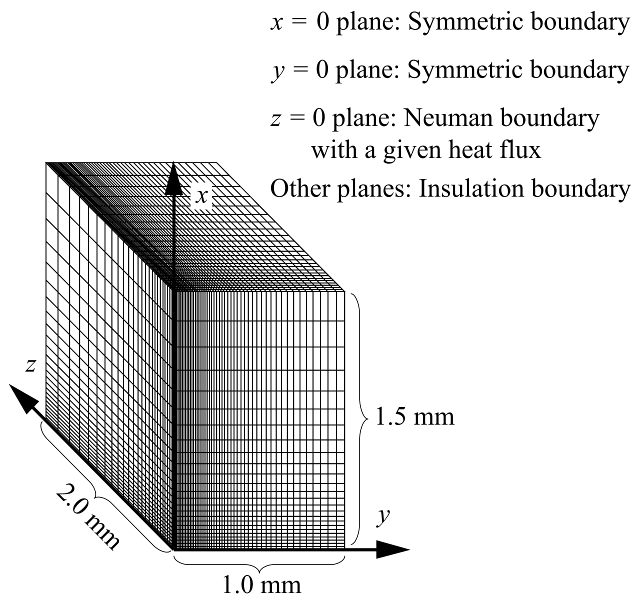

Fig. 4 Calculation grid and domain.

according to finite differential method. Discretization on the boundary was derived assuming a controlled volume method. Time marching was fourth order Runge-Kutta method. The time step Δt is selected to satisfy the stability condition of the difference equation for the diffusion equation [14].

4.2 Thermal properties of B/KNO_3

Table 1 shows thermal properties used in this calculation. The density was calculated from the size and weight of the pellets. The specific heat was calculated by the mass average of boron and potassium nitrate. Reaction threshold temperature and heat of fusion were sited from Yano's result [12), 13)], according to our boron fraction rate.

The thermal conductivity is an important parameter to predict thermal behavior. However, it is difficult to accurately predict thermal conductivity of mixture solid of two components. In spite of many researches on this field [15], there are several discrepancies of those results according to the thermal conductivity and mixture ratio. In this study, empirical formulae obtained by Kobayashi et al. was used to predict the thermal conductivity [15]. Thermal conductivity of mixture solid predicted by other authors and the comparison with those are shown in the elsewhere [16].

Table 1 Thermal properties of B/KNO_3.

Born weight fraction: p	0.30
Density: ρ	2230 kg m^{-3}
Specific heat: c_p	1140 J kg^{-1} K^{-1}
Thermal conductivity: k	1.6 W m^{-1} K^{-1} *
Reaction threshold: T_R	720 K
Heat of reaction: Δh_R	4.0 MJ kg^{-1}

*Thermal conductivity of mixture solid (boron: 27.6 and potassium nitrate: 0.5 W m^{-1} K^{-1}) calculated by the prediction equation.

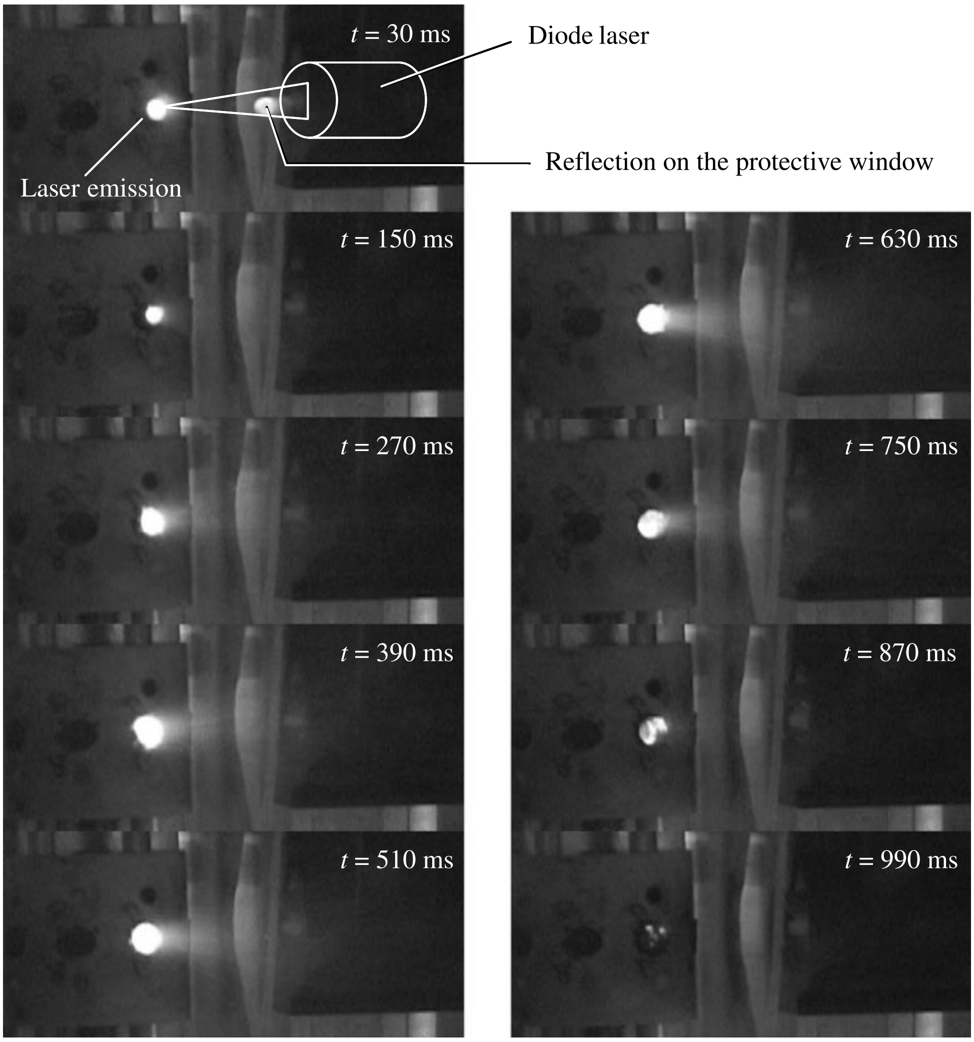

Fig. 5 Succeeding pictures of laser-ignited combustion of B/KNO_3 in vacuum.

5. Results and discussions

5.1 Experimental results of ignition probability

Figure 5 shows the succeeding pictures of a typical laser-ignited combustion (300 mW). The device used in the picture was prepared not for ignition probability examination but for micro space propulsion system [2], where the combustion processes can be well observed. Laser pulse width was set as 100 ms. Initially, the laser beam irradiates the pellet (t = 30 ms). The pellet then undergoes combustion at the focal point of the laser beam (t = 90 ms). The combustion front spreads over the surface of the pellet and steady combustion lasts during t = 270 - 690 ms.

The effect of laser power on the ignition probability was investigated. The measured ignition probability is shown in Fig. 6, which was obtained after the ignition trials using 80 pellets. There the ignition probability is defined as the number of ignitions over number of trials. In the experiment, laser pulse width was fixed at 1000 ms to remove the effect of laser pulse width. The threshold power of the laser ignition was estimated as 290 mW. No pellets was ignited under the 200 mW of power, and all pellets were combusted over the 400 mW. Curved line in Fig. 6 is a shifted error function, whose center point was determined by the fitting on the data.

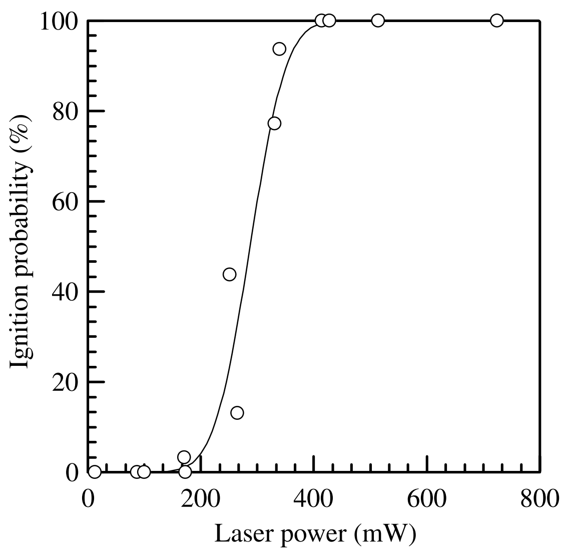

Fig. 6 Laser ignition probability of B/KNO_3 in vacuum, dependence on laser power.

5.2 Calculation results and their comparison with the experiment

Three dimensional numerical calculation was conducted with changing the laser power from 0.2 to 0.7 W. Te calculation was performed under several conditions to cover the uncertainties included in the vertical beam width and reaction rate; vertical laser width from 30 to 110 μm and reaction rate from 10 to 130 s^{-1}. As a result, three kind of typical results are obtained: no reaction, quenching, and self-sustained combustion. No reaction means any point does not reach to the threshold temperature within one second. Quenching means several points reach to and over the threshold, and exothermic reaction occurs, but that reaction cannot be sustained. In the self-sustained combustion, chain reaction of the exothermic reaction occurs, and the whole volume is reacted. Figure 7 shows the typical temperature profiles on y = 0. With the laser power of 0.40 W and the reaction rate of 30 s^{-1}, the maximum temperature reaches to 720 K at t = 50 ms and exothermic reaction starts.

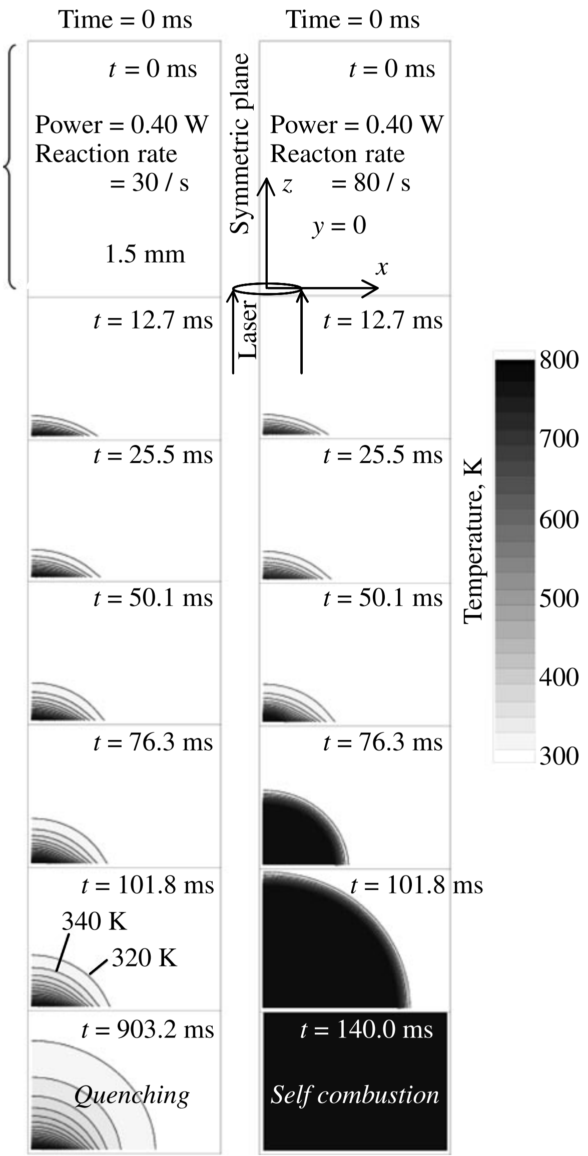

Fig. 7 Calculated temperature profiles on y = 0 plane; comparison of the quenching (reaction rate of 30 s^{-1}) and self-sustained combustion (reaction rate of 80 s^{-1}).

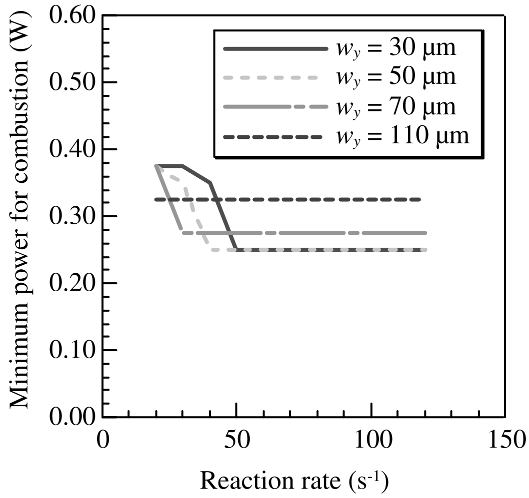

Fig. 8 Calculated minimum power necessary for the self combustion.

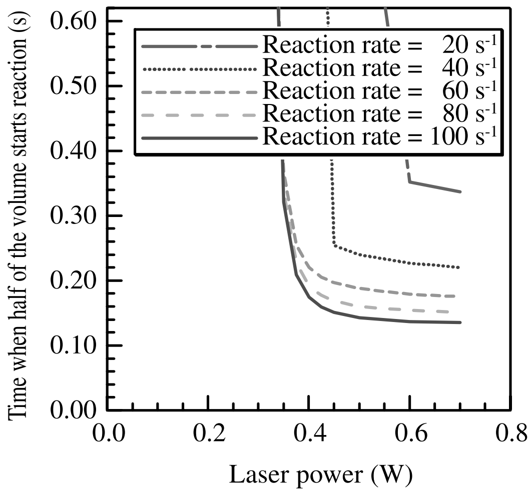

Fig. 9 Calculated ignition time when 55 % of volume starts the reaction (w_y = 50 μm).

However, after that the reaction did not grow any more. In the case of 80 s^{-1}, chain reaction is accomplished and the combustion propagates to whole volume.

Figure 8 shows the calculated minimum power necessary for the self-sustained combustion. The threshold power exists between 0.3 to 0.5 W. It well agreed with the experimental results of ignition probability measurement. In that experiment, we could not know the accurate vertical laser width of the laser because of the uncertainty of the pyrotechnics and laser beam. The associated position error would be less than ± 0.5 mm, and it causes the inaccuracy of laser width from 30 to 110 μm. However, it is shown by the calculation that the threshold power is changed only ±0.05 W corresponding to the width change from 30 to 110 μm.

Figure 9 shows the time when the reactive fraction of volume reaches to 55 % (w_y = 50 μm). With the enough laser power, the ignition times are ranged from 150 to 350 ms according to the reaction rate. Reaction rate more than 40 s^{-1} gives the ignition time around 200 ms. That value almost agrees with the experimentally observed combustion pictures.

6. Conclusion

In this study, we have performed the measurement of laser ignition probability in vacuum using a diode laser and the numerical calculation of the inner heat conduction with exothermic reaction. As a result we obtained the following conclusions.

(1) Boron / potassium nitrate can be ignited in vacuum using a diode laser. The threshold power necessary for the ignition was 290 mW and there was no miss-firing in the case of over 400 mW.

(2) The calculated threshold power and ignition time well agreed with the experimental results. In the calculation, laser beam width was changed from 30 to 110 μm, and there was no large change of the threshold power, only ± 50 mW difference.

References

1) S. Tanaka, R. Hosokawa, S. Tokudome, K. Hori, H. Saito, M. Watanabe, and M. Esashi, Trans. Jpn. Soc. Aeronaut. Space Sci., 46, 47 (2003).
2) H. Koizumi, T. Inoue, M. Nakano, and Y. Arakawa, J. Propul. Power, 21, 1133 (2005).
3) M. Nakano, H. Koizumi, T. Inoue, M. Watanabe, and Y. Arakawa, 25th International Symposium on Space Technology and Science, ISTS 2006-b-05 (2006), Kanazawa.
4) K. Takahashi, T. Okada, T. Ikuta, T. Nishiyama, and K. Nagayama, 25th International Symposium on Space Technology and Science, ISTS 2006-b-03 (2006), Kanazawa.
5) N. K. Bourne, Proc. R. Soc. Lond. A, 457, 1401 (2001).
6) L. D. Yong and F. Lui., Prop., Explos., Pyrotech., 23, 328 (1998).
7) C. Zanotti and P. Giuliani, Prop., Explos., Pyrotech., 23, 254 (1998).
8) F. Opdebeck and P. Gillard, Prop., Explos., Pyrotech., 26, 196 (2001).
9) F. Opdebeck, P. Gillard, and E. Radenac, Int. J. Therm. Sci., 42, 47 (2003).
10) S. R. Ahmad and D. A. Russell, Prop., Explos., Pyrotech., 26, 235 (2001).
11) D. Ding, Sci. Tech. Energetic Materials, 67, 23 (2006).
12) Y. Yano, Prop., Explos., Pyrotech., 14, 187 (1989).
13) Y. Yano, J. the Industrial Explosives Society (Sci. Tech. Energetic Materials), 49, 129 (1988).
14) K. Fujii, “Numerical methods for computational fluid dynamics”, p. 35 (1994), University of Tokyo Press.
15) M. Kobayashi, H. Maekawa, H. Nakamura, and Y. Kondou, Trans. Jpn. Soc. Mech. Eng. B, 57, 271 (1991).
16) H. Koizumi, “Study on Micro Space Propulsion”, Ph. D. Dissertation, Univ. of Tokyo (2006).

真空中におけるボロン／硝酸カリウムのレーザー着火に関する研究

小泉宏之 *†，中野正勝 **，井上孝祐 *，渡辺将史 ***，小紫公也 ****，荒川義博 *

本研究では真空中におけるボロン／硝酸カリウムの着火特性を半導体レーザーを着火エネルギー源として用いた場合に関して明らかにした。実験によって真空中のレーザー着火確率を測定した結果，着火に必要な最低パワーは0.4 Wであった。また，火薬内部の発熱反応を考慮し熱伝導の数値解析を行い，実験結果との比較を行った。その結果，着火閾値パワーをはじめとする両者の結果は良好に一致することが示された。

* 東京大学　工学系研究科　航空宇宙工学専攻　〒113-8656　東京都文京区本郷 7-3-1
† Corresponding address: koizumi@al.t.u-tokyo.ac.jp

** 東京都立航空工業高等専門学校　〒116-8523　東京都荒川区南千住 8-17-1

*** 日油技研工業株式会社　〒350-1107　埼玉県川越市的場新町 21-2

**** 東京大学　新領域創成科学研究科　先端エネルギー工学専攻　〒277-0882　千葉県柏市柏の葉 5-1-5