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Ignition time of self-propagating high-temperature synthesis by laser ①

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[Abstract] The ignition of self-propagating high-temperature synthesis (SHS) by a laser beam has very well application, but there is lack in study on the ignition process. In order to search the rule of ignition process with laser beam, ignition time of SHS was studied in detail. First one dimension ignition model was introduced: burning is the process in which one layer is ignited by next layer. Then according to Fourier conduction equation, an equation used to calculate the ignition time was deduced. Finally a series of tests were made to verify the equation. The results prove that the change of the parameters in test agrees well with the equation.

[Key words] SHS; laser; ignition model; ignition time

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1 INTRODUCTION

SHS, the self-propagating high-temperature synthesis, was first discovered by Merzhanov and Borovinskaya when they studied solid fuels of solid rocket. SHS begins to burn continuously until all reactive material have finished as soon as an exothermic heat of the chemical reaction is ignited by the external heat source. SHS has unique advantages: 1) simple process, shorter reaction time; 2) more pure synthesis; 3) saving energy; 4) material synthesis and many other kinds of processes such as sintering, welding etc can be integrated into one process[1~4].

Ignition is a very important process of SHS. Gasless system plays an important rule in synthesis and making of the material of SHS, but the research on its ignition is not enough[5].

For ignition model, the energy resource which gives SHS material enough energy to rise its temperature to reach ignition point may be used theoretically to ignite the SHS. Nowadays the common ignition sources are as follows. Tungsten wire like dish[5], contact resistance[6], welding arc[7], microwave[8], thermal explosion[9], shock loading[10], laser beam[11~14]. There are two kinds of lasers used in ignition: pulse[11] and continuous[12~14] laser beam.

The temperature of synthesis reaction, the heat flux, the synthesis reaction process and the synthesis have been mainly studied after igniting by different kinds of laser beams. The results of the research completed prove that burning procedure and synthesis have no essential difference between laser ignition and common source ignition[11~14], but the most difference is in ignition time. Up to now there is still not detailed study about this stage.

The controlling of ignition SHS by laser beam is mainly at the ignition process, so the ignition process with laser beam is investigated in detail in this paper. First an equation is reduced according to the one dimension (1-D) model introduced, then a series of experiments are done to verify the equation.

2 IGNITION MODEL AND EQUATION

2.1 Ignition model of SHS

The gasless system of SHS 1-D ignition model is introduced from Ref.[5] as shown in Fig.1. First layer is ignited by external ignition source, the layer next to the first one is then ignited by the heat released by the reaction of the first layer. So the ignition procedure happens from layer to layer until SHS finishes.

Laser beam which fast scans back and forth on the sample surface along scanning line may imitate line heat source to ignite SHS as first process of the 1-D model.

2.2 Deducing equation of model

The basic assumptions for the 1-D model are as follows from Ref.[5]:

1) Temperature is unique in the cross section of sample;

2) Physical parameters do not change along with its temperature;

3) The sample is isolated from surroundings.

Let starting temperature be T_2 as shown Fig.1.

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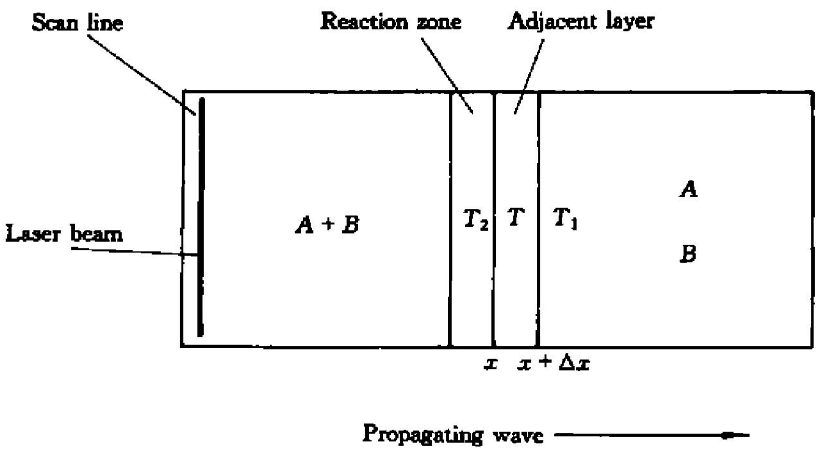

Fig. 1 Schematic representation of gasless SHS process

The thickness at unreaction layer next x is Δx and the temperature is T. The temperature is T_1 at $x+\Delta x$. According Fourier basic heat conduction equation for 1-D model there is:

$$\frac{\partial^2 T}{\partial x^2}=\frac{\rho c}{K}\cdot\frac{\partial T}{\partial t}=\frac{1}{\alpha}\cdot\frac{\partial T}{\partial t} \tag{1}$$

where ρ is density, c is thermal capacity, K is thermal conductivity, t is time, and α is temperature conductivity.

According Ref. [5] a parameter is introduced:

$$Y=\frac{x}{2\sqrt{\alpha t}}$$

The integral upper limit α and low limit b can be obtained by following boundary conditions:

$$x\to 0,\ t\to 0,\ a\to 0$$

$$x\to x,\ t\to t,\ b\to\frac{x}{2\sqrt{\alpha t}}$$

Thus

$$T=C_1\int_0^{\frac{x}{2\sqrt{\alpha t}}}\exp(-Y^2)\,\mathrm{d}Y+C_2 \tag{2}$$

According definition and assumption, we have

$$\lim_{t\to\infty}T=T_i \tag{3}$$

where T_i is the ignition temperature.

Then, we can get

$$C_2=T_i \tag{4}$$

and

$$\lim_{t\to 0}T=T_a \tag{5}$$

where T_a is the starting temperature.

We can also get:

$$C_1=\frac{2(T_a-T_i)}{\sqrt{\pi}} \tag{6}$$

and

$$T=\frac{2(T_a-T_i)}{\sqrt{\pi}}\int_0^{\frac{x}{2\sqrt{\alpha t}}}\exp(-Y^2)\,\mathrm{d}Y+T_i \tag{7}$$

To solve Eqn. (7), and then the derivation of it with respect to x, we get the temperature gradient:

$$\frac{\partial T}{\partial x}=\frac{T_a-T_i}{\sqrt{\pi\alpha t}}\exp(-x^2/4\alpha t) \tag{8}$$

According the definition and Eqn. (8), 1-D heat flux equation is

$$q=-KS\frac{\partial T}{\partial x}=KS\frac{T_i-T_a}{\sqrt{\pi\alpha t}}\cdot\exp(-x^2/4\alpha t) \tag{9}$$

where q is heat flux conducted into next layer, and S is the cross section.

When heat conduction reaches an instantaneous equilibrium process at x, its temperature stops changing. Let

$$\lambda=\exp(-x^2/4\alpha t) \tag{10}$$

where λ is a constant.

Put Eqn. (10) into Eqn. (9), then

$$q=\frac{KS\lambda(T_i-T_a)}{\sqrt{\pi\alpha t}} \tag{11}$$

According to the model, laser ignition is the first ignition process, so we have

$$q_i=q \tag{12}$$

where q_i is heat energy absorbed by metal powder.

A part of laser beam energy is absorbed and changes into heat energy, another part is reflected.

Heat energy absorbed by the surface of sample is

$$q_i=(1-R)q_a=4.186(1-R)P \tag{13}$$

where q_a is total heat energy of laser beam, R is reflective coefficient, and P is power of laser beam.

When laser beam radiates vertically, the reflective coefficient is[15]

$$R=\frac{(n-1)^2+k^2}{(n+1)^2+k} \tag{14}$$

where n is index of refraction, and k is index of absorption.

Put Eqn. (14) into Eqn. (13), we get

$$q_i=4.186\left[1-\frac{(n-1)^2+k^2}{(n+1)^2+k}\right]P \tag{15}$$

Put Eqn. (15) and Eqn. (11) into Eqn. (12), we get

$$4.186\left[1-\frac{(n-1)^2+k^2}{(n+1)^2+k}\right]P=\frac{KS\lambda(T_i-T_a)}{\sqrt{\pi\alpha t}} \tag{16}$$

Finally, the ignition time equation is

$$t = \left[\frac{KS\lambda(T_i - T_a)}{4.186P\sqrt{\pi\alpha}} \cdot \frac{(n+1)^2 + k}{4n + k - k^2}\right]^2 \quad (17)$$

Eqn. (17) is used to calculate the ignition time. Then, a series of experiments is done under the change of the changeable parameters in Eqn. (17) to verify the equation.

3 EXPERIMENTAL

3.1 Experimental design

There are two factors to decide the ignition time. One is energy density of ignition source. Density of laser beam is many times larger than that of common ignition sources, but only a small part of total laser beam energy is absorbed, most of them is reflected. Absorption capacity depends on many factors, such as: nature of material, the material with good conductivity has bad absorption capacity; surfaces of samples with roughness and materials with high melting point have good absorption capacity. SHS material is made up of several kinds of materials, and its absorption capacity is unified absorption capacity of several kinds of materials. High roughness of the sample's surface benefits its absorption capacity. The other is the material conductivity. A sample made up of metal powder with higher density has better conductivity, so it needs longer time to ignite the sample.

Eqn. (17) is verified by tests of all kinds of factors that effect the two aspects above.

3.2 Experimental methods

Test device is shown in Fig. 2. The laser beam produced by CO_2 laser passes through an optical system and dynamic scanning system to be projected on the sample surface. Laser beam scans back and forth on the surface at one end of the sample; maximum output power of laser is 50 W.

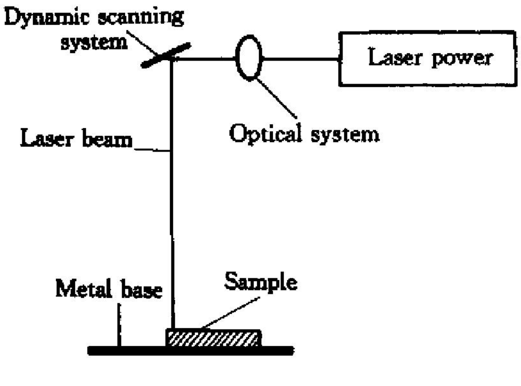

Fig. 2 Scheme of experimental set up

Common test parameters are as following except a special parameter of every test.

Output power of laser is 40 W, spot size diameter is 0.12 mm, scanning distance is 20 mm, scanning speed is 200 mm/s, sample made up of metal powders is formed at nature state without any press, and test is done in air.

The main test material is Ni-Al, Al-Ti and Ni-Ti are seldom tested. Ni, Al, and Ti metallic powders are used in industry. The particle size of Ni and Ti is 74 μm, that of Al is 50 μm, and the theoretical fraction of two kinds of material are 1∶1 for both Ni-Al, Al-Ti and Ni-Ti. The mass of sample is 10 g and its gauge is 45 mm × 20 mm × 10 mm. Fig. 3 is the photo of samples after SHS.

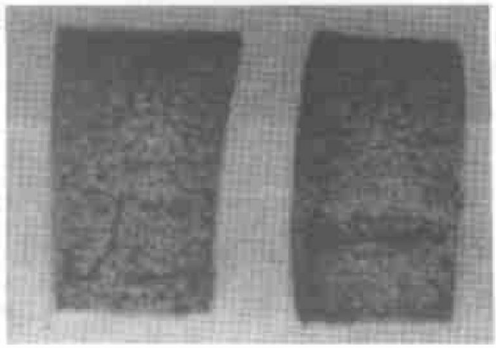

Fig. 3 Photos of samples

The ignition time is from the laser beam striking on the sample's surface to the burning wave reaching place which is 5 mm distance from the scanning line, because burning wave out of 5 mm becomes self-propagating wave.

4 RESULTS AND DISCUSSION

4.1 Ignition time of laser power

First we need to decide the basic time to mix the materials. All the test materials weighted are filled into the mixer at the same time to mix. Test material needed is fetched from the mixer at the time specified, and the rest materials keep mixing for getting exactness results.

Ignition time becomes shorter as the mixing time extends because the material is more unique, but over certain time, ignition time becomes longer with growth of mixing time on the contrary. Its reason is oxiding of Al. Ignition time is the shortest when the mixing time is 0.75 h and the output power is 40 W at room temperature.

The shorter the ignition time is, the larger the laser power goes (see experiment curve in Fig. 4).

From Eqn. (17) we know that the square of ignition time has a hyperbola relation with laser power (see theorectical curve in Fig. 4). The experimental result is a near hyperbola curve, so they have a same relationship.

4.2 Ignition time of different pre-heat temperatures

The sample is pre-heated. As soon as the temperature reaches the required point, the ignition immediately starts by the laser beam.

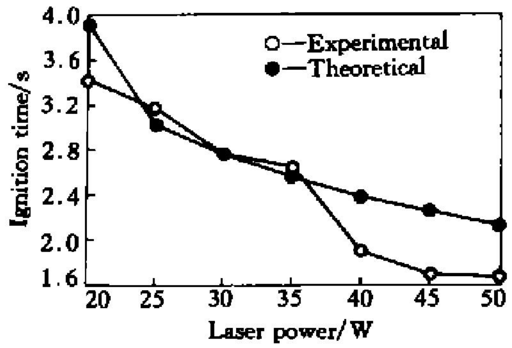

Fig. 4 Relation of ignition time with laser power

Ignition time becomes shorter with increasing temperature. There are two reasons, one is that increasing of material temperature makes growth of absorption capacity, and leads to adding of effective energy heated, and the other is that pre-heat adding input of heat (see experimental curve in Fig. 5).

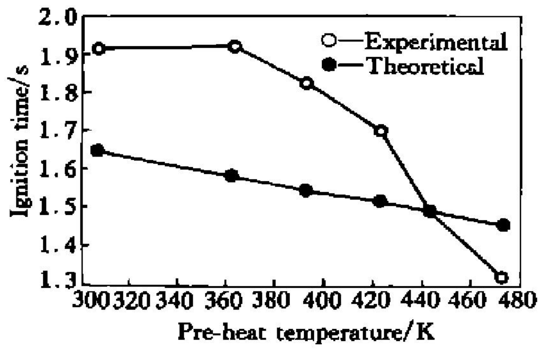

Fig. 5 Relation of ignition time with pre-heat temperature

The pre-heat temperature is starting temperature, from Eqn. (17) we have it as follows:

$$t = \sqrt{\frac{T_i - T_a}{C}} \quad (18)$$

or

$$T_a = T_i - Ct^2 \quad (19)$$

where C is a constant.

Eqn. (19) is a parabola with its mouse towards terra (see theoretical curve in Fig. 5). From Fig. 5 we know that the experimental curve is also a part of a parabola with its mouse towards terra, but the curvature of theoretical curve is larger than that of experimental curve. They have a same relationship.

From Fig. 5, we also know that there is clear error between the experimental curve and theorectical curve. The reason causing this case is that the theorectical model is 1-D. In fact, the combustion reaction happens in 3-D model.

4.3 Ignition time of different kinds of materials with different melting points and different scanning speeds

Three kinds of materials with different ignition points and near absorption capacity were tested. It is impossible for Ni-Al to ignite when scanning speed was over 500 mm/s.

Ignition time increases with increasing ignition point (the ignition point of Ni-Al is about 1073 K[16], Al-Ti is 1100 K, Ni-Ti is 1423 K[17]) (see Fig. 6). According to Eqn. (17), T_a increases and ignition time becomes longer.

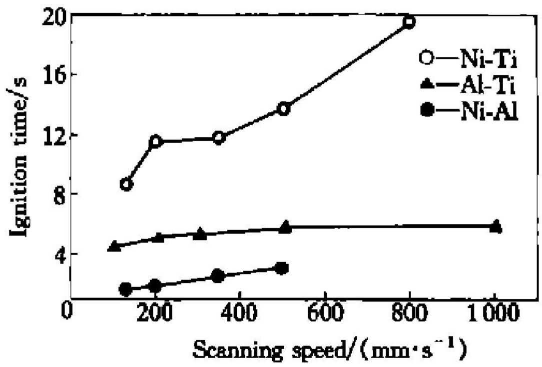

Fig. 6 Relation of ignition time with scanning speed

The ignition time increases with increasing scanning speed (see experimental curves in Fig. 6). The energy density that is used to heat the sample decreases with increasing the scanning speed when the spot size is not change.

4.4 Ignition time of different densities

A lubricant is not used during forming sample for keeping precision, but it is helpful for the sample to be broken as the density of the sample is less than 2.428 g/cm^3, as the density is 3.528 g/cm^3, the sample is not ignited in 180 s.

Ignition time increases along with increasing density of the sample, the causing reason is the increasing of conductivity (see Fig. 7). From Eqn. (17) we know that ignition time has a direct radio relation with the thermal conductivity. The thermal conductivity increases with increasing density of the sample.

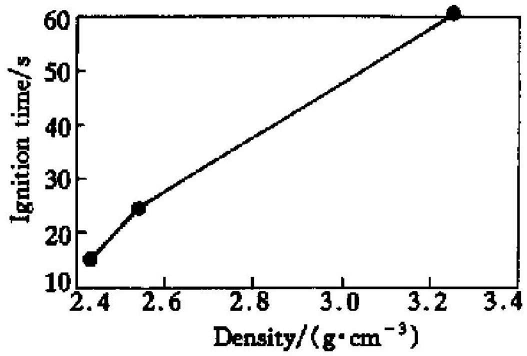

Fig. 7 Relation of ignition time with density

4.5 Ignition time of different spot size diameters

Ignition time increases with increasing spot size diameter of laser beam (see experimental curve in Fig. 8).

The experiment curve has a same trend with the theoretical curve in Fig. 8.

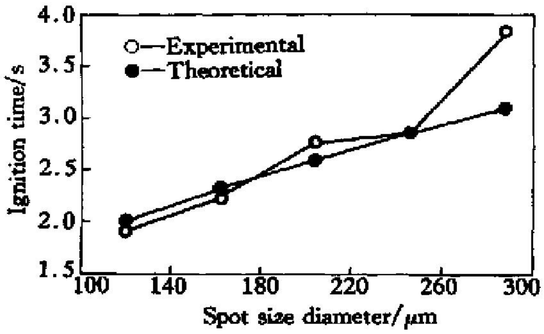

Fig. 8 Relation of ignition time with spot size diameter of laser beam

5 ANALYSES

5.1 Density of energy

There are two methods for changing the density of energy, one is only changing the output power of laser, the other is changing scanning speed with constant output of laser. Two kinds of tests have the same result: ignition time decreased with increasing density of energy, which coincides with Eqn. (17).

5.2 Pre-heat

Ignition time decreases with increasing pre-heat temperature, because starting temperature can be increased by pre-heat. Eqn. (17) and the test have the same results.

5.3 Conductivity

The conductivity increases when the sample's density increases, the ignition time increases, which coincides with Eqn. (17).

5.4 Spot size diameter of laser beam

The ignition time increases with increasing spot size diameter. Both equation and experiment results have the same result.

5.5 Different ignition temperature

Ignition time rises with increasing ignition point, and this result is as same as the result of growth of T_i in Eqn. (17).

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