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ADVANCED OPTICAL DIAGNOSTIC TECHNOLOGY APPLICATION PROGRESS IN COMBUSTION TESTING OF ENERGETIC MATERIALS

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Abstract: Based on different optical principles, laser-induced fluorescence (LIF), coherent anti-Stokes Raman scattering (CARS), particle imaging velocimetry (PIV), and tunable are reviewed from three aspects: light scattering, optical emission and absorption, and imaging. Testing principles of optical diagnostic technologies such as semiconductor laser absorption spectroscopy (TDLAS), laser-induced breakdown spectroscopy (LIBS), radiation method, remote sensing Fourier transform infrared spectroscopy (RS-FTIR) and schlieren method, and their use in combustion tests of energetic materials The application progress of optical diagnostic technology in combustion testing is analyzed. The superiority of optical diagnostic technology compared with other traditional contact diagnostic methods and the applicability, measurement objects, advantages and disadvantages of various optical diagnostic methods are analyzed; the microscopic combustion products, flame temperature, The development prospects of testing technologies such as combustion flow field velocity and flame structure in combustion diagnosis of energetic materials; it is pointed out that future work should combine multi-diagnostic methods and develop multi-dimensional measurements to obtain richer and multi-dimensional microscopic data information.

Keywords: Physical chemistry; Combustion of energetic materials; Optical diagnostic technology; Laser-induced fluorescence; Laser absorption spectroscopy; Schlieren method

1 INTRODUCTION

The control and regulation of the combustion properties of energetic materials are of great significance for efficient energy release and meeting the application needs of different weapons and equipment. Different from the combustion of other fuels, the combustion of energetic materials is generally more complex. During combustion, it can produce high temperature and high pressure, and undergo a series of complex physical and chemical changes from solid phase to liquid phase or gas phase. The combustion environment is usually accompanied by high pressure, vibration and other external factors. factor. Solid propellant produces soot particles during the combustion process, and the burning surface of metal-containing propellant is accompanied by agglomeration of metal particles and other phenomena. Different from conventional gas or liquid fuels, energetic materials themselves contain oxidants and combustibles, and can independently carry out redox reactions under certain external stimuli. They can burn under anaerobic conditions and release a large amount of gas and heat. Regarding the combustion behavior of energetic materials, although researchers have established many relevant combustion models, there is still a lack of clearer understanding of the combustion of energetic materials with different formulas and the flame structure and reaction mechanism of combustion in the combustion chamber of high-pressure rocket engines. The development of theoretical simulations lacks the new full range of information provided by experiments. In order to obtain the essential laws of the combustion process, understand the detailed dynamics of the combustion reaction and the changing characteristics of the combustion flow field, reveal the combustion mechanism of energetic materials from a microscopic perspective and adjust the combustion efficiency, obtaining combustion information needs to be microscopic, systematic, and real-time online.

With the continuous development of combustion diagnostic technology, traditional contact experimental diagnostic methods, such as thermocouple temperature probes, gas sampling probes, etc., will inevitably cause disturbances to the combustion flow field. The complex combustion test environment of energetic materials also limits The accuracy and application range of the probe have been reduced, and it has gradually become difficult to meet the needs for more effective fast online real-time measurement. Non-contact optical diagnosis methods, because they only require the detection of flame emission or absorption characteristic spectral information, reduce or avoid aerodynamic, thermal or chemical disturbances, can withstand high temperatures and harsh environments, and have higher spatial and temporal resolution.

This article distinguishes based on the principles of optical diagnosis. From the three aspects of light scattering, optical emission and absorption, and imaging, the laser-induced fluorescence (LIF) and coherent anti-Stokes Raman scattering methods in recent years are distinguished. (CARS), laser absorption spectroscopy (LAS), laser-induced breakdown spectroscopy (LIBS), radiation method, remote sensing Fourier transform infrared spectroscopy (FTIR), particle imaging velocimetry (PIV) and schlieren method in energy-containing The applications in materials and engine combustion diagnosis are reviewed, and the advantages and disadvantages of specific methods in realizing real-time detection of information such as combustion flame temperature, flame structure, product component concentration, and combustion flow field velocity under different environmental conditions are analyzed.

Prospects for future diagnostic technologies and needs in the field of energetic material combustion are proposed.

Table 1 Common advanced optical diagnostic methods for energetic material combustion

principle	Classification	name	application
light scattering	First-order inelastic scattering Third-order nonlinear scattering	LIF, PLIF CARS	Detect group components such as NO, OH, CH, CN and metal atoms such as Fe and Al. Flow field temperature measurement detects CO and N2., H2 Isomolecule and flow field temperature measurement
Optical emission and absorption	emission	radiation method	Combustion flow field temperature measurement and flame structure
		RS-FTIR	Remotely measure flame temperature
		LIBS	Qualitative and quantitative analysis of non-metals such as C and O and metallic elements such as Al and Ir
	absorb	LAS	CO, CO2 and other molecular concentrations and temperature measurement
light imaging		PIV	Combustion flow field velocity measurement (overall)
		schlieren	Density gradient, combustion flow field changes

2 LIGHT SCATTERING MEASUREMENT

The energy is hv0 When a multi-phase combustion flow field composed of molecules and particles is irradiated by a laser, light absorption is not considered, and the scattered light is classified according to the emission frequency, which can be divided into first-order elastic scattering processes such as Rayleigh scattering and Mi(Mie) scattering, first-order inelastic scattering processes such as laser-induced fluorescence (LIF), and third-order nonlinear scattering processes such as coherent anti-Stokes Raman scattering (CARS). Light scattering method is essentially a spectroscopic method, which uses the analysis of spectral signals as the basis for diagnosis. The following is a review of LIF and CARS technologies that are widely used in the field of energetic materials and have great development prospects.

2.1 Laser-Induced Fluorescence (LIF)

Laser-Induced Fluorescence Fluorescence (LIF) is the use of a frequency-adjustable laser to generate laser light to irradiate atoms or groups, causing them to transition from a metastable state to an excited state. Since the excited state is not stable, it will spontaneously radiate energy to a lower energy level and return to the ground state., the energy generated during the process is released in the form of fluorescence, and the fluorescence signal is then detected and analyzed by the testing technology (see Figure 1). In principle, it is a first-order inelastic scattering process, and the fluorescence survival time is 10 -10 ~10-5 s. LIF technology can measure one-dimensional component concentration, temperature and other parameters of the combustion environment in real time, and has good spatial resolution, and can measure OH and NO, etc. 10 -6 amount of active ingredient. The LIFs of different components and quantum states are different and specific. There is a functional relationship between the concentration of the component and its fluorescence signal intensity. Using a laser with a specific frequency of a certain component for excitation, and collecting the signal of the corresponding frequency can complete the concentration of the target component. Therefore, PLIF technology is used in online diagnosis of combustion product concentration and flame structure [1-4] It has a wide range of applications: analyzing the changes in combustion wave structure by measuring the concentration and spatial distribution of free radicals, thereby helping to understand the combustion mechanism of energetic materials. This technology has become an important experimental diagnostic tool for engine combustion and propellant combustion flames.

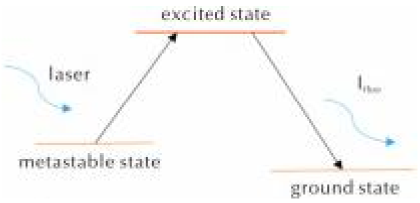


Figure 1 Schematic diagram of the principle of laser-induced fluorescence

The application of LIF technology in the field of energetic material combustion was originally used by Edwards et al. to burn Octogen (HMX) and ammonium perchlorate/hydroxy-terminated polybutadiene (AP/HTPB) composite propellants at 3.5MPa. Experimental diagnosis. Later Parr et al[5] LIF was applied to other types of solid propellants, the LIF signals of several free radicals (such as CN, NH, NO, OH) were monitored, and the combustion characteristics of HMX/GAP/BTTN solid propellants were studied using OH-LIF and other technologies. The flame structure, combined with OH-LIF, UV/vis data and thermocouple measurements, more accurately corrects the burning surface temperature and flame zone temperature during solid propellant combustion. Before the LIF excitation light enters the combustion flow field, it is first formed into a sheet laser through the sheet light shaping system, which is developed into Planar Laser-Induced Fluorescence, PLIF) technology, because PLIF technology can obtain two-dimensional information parameters of the combustion environment, it is more widely used in energetic material combustion and flow field diagnosis research in engine combustion chambers.

Yan Zhiyu et al[6] Using OH-PLIF, NO chemiluminescence and high-speed photography methods, the self-luminescence process of RDX reaction and the OH inside the flame before and after focused ignition were studied. Two -dimensional concentration distribution of free radicals, RDX is briefly analyzed based on the results combustion mechanism. Ruesch et al. [7] used PLIF to study CN in hexanitrohexaazaisowurtzitane (CL-20) cocrystal and HMX/AP polycrystalline composite crystals. and OH concentration distribution and flame structure during the combustion process, and the obtained PLIF flame structure changes were used to explain the difference in combustion rates between HMX/AP physical mixtures and composite crystals. Kevin et al[8-9] This system was used to study the combustion process of gel propellant droplet jets. At a time resolution of 0.2ms, three different types of jets were observed. This two -dimensional visualization of the flame structure of gel jets based on PLIF enables Scholars have a more basic understanding of the combustion laws of gel fuel droplets.

The planar visualization technology provided by PLIF satisfies the diagnosis of flame structure at the two-dimensional level, but for the three-dimensional measurement of the complete structure of the flame in three-dimensional space, the development of new testing technology is still needed. Peterson et al. [10-11] used two sets of PLIF systems to study the flame propagation changes in the early stages of engine ignition through OH concentration distribution. The system realized the PLIF signal tomographic inversion of OH. three-dimensional spatial distribution. Yuan Xun et al. [12] designed and built a multi-plane 3DLIF system based on scanning galvanometers to meet the needs of supersonic flame combustion diagnosis and the limitations of high-frequency scanning technology, which can perform high-frequency scanning in the 20mm sheet light range and realize OH-3DLIF spatial visualization.

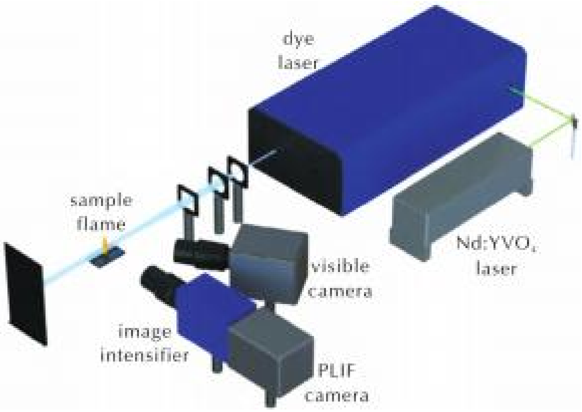


Figure 2 Schematic diagram of PLIF test system[7]

In addition to the determination of combustion product concentration distribution and flame structure, LIF The technology is also widely used in combustion flow field temperature measurement. Currently, the most commonly used method for LIF temperature measurement is the double-line method. During temperature measurement, two laser beams of different wavelengths need to be used to excite two different rotational energies of a certain group at the same vibration energy level at the same time or within a very short time. level transition line, and then calculate the flame temperature through the ratio of the obtained fluorescence signal intensity at the two wavelengths. The test system is shown in Figure 3. This method can avoid the influence of free radical concentration and fluorescence quenching effect to a certain extent. Since OH contains abundant excitation spectral lines in the combustion reaction zone and high temperature zone, OH is often used as the laser action medium for temperature field measurement.. Due to the strong dependence of OH concentration on temperature, OH-LIF temperature measurement is generally suitable for T > 1300K combustion environment[13].

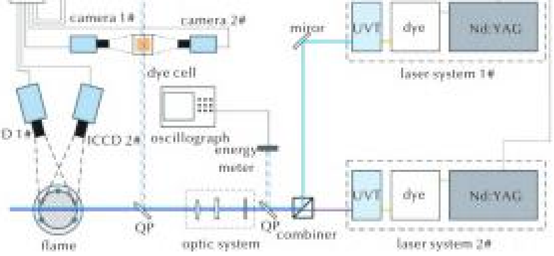


Figure 3 Two-wire PLIF temperature measurement system

Foreign research on LIF dual-line temperature measurement was earlier. In 1984, Soloukhin et al. from Sandia Laboratories in the United States[14] First use to describe the density of absorption states [15] The Boltzmann population function relates the ratio of fluorescence signals to temperature, and uses the ratio of the two fluorescence image intensities to determine the temperature distribution of the flame. This has also become a classic solution for LIF temperature measurement since then. Lu Yongxu[16] used dual-line OH-PLIF technology to measure the two-dimensional temperature distribution of the combustion flame of propellant samples, and combined with the results of thermocouple calibration experiments, determined Q2 (11) and P1 (7) The two excitation spectral lines are the best spectral line selection for the dual-line PLIF technology to measure the alcohol lamp flame and the combustion flame temperature of a certain type of propellant, which provides ideas for the testing method of formulated propellants. There is also dual tracer temperature measurement, which uses two tracers and tests with two different excitation frequencies or with lasers with the same excitation frequency. This method follows the principle of dual-line temperature measurement and uses the fluorescence intensity ratio to measure temperature. Measurement calculations.

Because aluminum atoms have strong fluorescence characteristic signal intensity, it produces a good signal-to-noise ratio [17-20], improves the LIF sensitivity in solid propellant flames, and the development of aluminum-based LIF enables a more accurate optical test method for measuring the combustion temperature of aluminum-containing solid propellants. Gas-phase AlO plane laser-induced fluorescence technology has been applied to single particle aluminum combustion temperature and AlO concentration [21] Under measurement. Chevalier [22] carried out a combustion experiment of aluminum-containing solid propellant at a pressure of 1.0MPa, using high-speed Al-LIF to detect the generated aluminum vapor, and using a one-dimensional quasi-steady state model to simulate the concentration and temperature of aluminum atoms around the combustion of aluminum droplets. distributed. The comparison shows that the experimental data and simulation results are consistent, indicating that the Al-LIF method can provide verification data for building a more accurate aluminum combustion model. Vilmart[23-24] AlLIF is used in high temperature applications (1200K and 2600K) and medium to low pressure (100Pa to 0. 1MPa) environment, a model of the relationship between Al-LIF spectrum and temperature was established, and the model was used to estimate that the LIF signal of Al can still be detected under high temperature and high pressure conditions (3000K and 10bar), which is Al The application of LIF-based in higher temperature environments provides theoretical support. For solid propellants containing aluminum, the combustion of aluminum particles increases the temperature and makes the flame highly luminous. The radiation background interference makes it difficult to accurately measure the combustion temperature by passive spectroscopy methods such as optical emission and absorption, which also reflects the advantages of the LIF method. Advantage. In addition, in the current study, iron-containing catalysts (ferrocene derivative-type catalysts) present in solid propellants can also be studied using LIF. Current experiments show that iron atoms have high fluorescence yield and high sensitivity, and the fluorescence signal can be easily detected. Vilmart et al. [25] conducted a Fe-PLIF-based study on AP/HTPB propellant combustion and established an iron atom LIF signal model, which further confirmed the real-time diagnostic capability of iron atoms in solid propellant combustion. In the future, microscale flow and phase changes on the solid propellant combustion surface, and the impact of additives on combustion performance (velocity, propellant surface behavior) are expected to be realized relying on metal atom PLIF technology and high-speed PLIF technology.

As an active non-contact optical diagnostic technology, PLIF has received more and more attention in solid propellant combustion diagnosis. Compared with other flow field diagnostic technologies such as schlieren technology mentioned later, PLIF has higher spatial resolution and more Low laser scattering effect on the flow field surface and high fluorescence yield of key combustion intermediates. The introduction of PLIF tracer molecules enables it to participate in the study of supersonic and hypersonic boundary layers. And the above-mentioned combustion intermediates almost have a common optical feature: their spectral transitions from the ground state to the first several excited states are in the ultraviolet wavelength range, which can be easily achieved using dye lasers or optical parametric oscillators (OPO). However, energetic materials still face some challenges: such as the difficulty of extracting LIF signals from strong laser scattering of condensed matter particles, high-pressure environments and continuous background emission, the attenuation of strong laser beams and the capture of fluorescence signals in dense media., and how to solve the pressure-dependent quenching of the LIF signal of free radicals to obtain quantitative information. Currently, PLIF technology has developed to 100KHz~1MHz, which can meet the temperature measurement requirements of medium Reynolds number subsonic turbulence with spatial and temporal resolution. However, it is still difficult to capture the detonation or transient combustion of hypersonic (1500m/s) flow that may occur in energetic materials.. Considering the instability of the flame, current combustion wave diagnosis requires increasingly higher time resolution, which also requires PLIF to develop towards higher frequency technology to meet the transient diagnosis of ultrasonic flow fields.

2.2 Coherent Anti-Stokes Raman Scattering (CARS)

Coherent Anti-Stokes Raman Scattering (Coherent AntiStokes Raman Spectroscopy (CARS) technology is a nonlinear four-wave mixing process. The pump light (ω1) and the Stokes light (ω2) are selected according to the measured molecular Raman shift (ωR). The two laser beams are phase matched. Incident into the flame area, the detection light (ω3) produces coherent light (ω4), which is the CARS signal, after mixing. Since the frequency difference between the pump light and the Stokes light is exactly equal to the Raman shift of the selected medium, the CARS signal intensity is greatly enhanced (see Figure 4). Due to the coherence properties, the CARS signal has laser-like characteristics and is almost unaffected by fluorescence and background light to obtain high spatial and temporal resolution and high-precision measurement results. This nonlinear optical scattering method will not interfere with the flame or flow field, and has played an important role in obtaining parameter information such as combustion field temperature [26-28] and component concentration [29-30]. Zhang Lirong et al. [31] used a self-developed integrated CARS system to measure the temperature of the exit jet of the supersonic combustion chamber, and obtained the single-pulse nitrogen CARS temperature fitting results and the temperature change model with time. The integrated CARS diagnostic system has good anti-vibration performance and can match on-site test conditions in a targeted manner, realizing the flexibility of CARS temperature measurement.

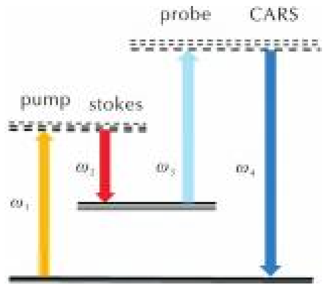


Figure 4 Schematic diagram of the basic principles of CARS

CARS in energetic materials that do not contain metal components [32-33] Chinese applications are relatively mature, including RDX Decomposition[34], SGP-38 [35], nitramine [36-37] and hydrazine nitroformate [37] combustion under different pressure conditions.

Hu Zhiyun et al. [38] used single-pulse CARS technology to diagnose the transient temperature and nitrogen concentration of the solid propellant combustion field under normal pressure and high pressure, and combined the theoretical calculation and fitting values to give the transient temperature of the solid propellant combustion field (about 2250K). and the distribution of nitrogen concentration with flame height. The results show that the broadband CARS technology can be applied to real-time diagnosis of solid propellant combustion field temperature under normal pressure and high pressure.

Traditional CARS technology uses high-energy pulsed nanosecond lasers with a repetition rate of 10Hz. In recent years, with the development of optical technology, femtosecond (fs) CARS with a repetition rate of up to 5kHz has been widely used in laboratory burners. The high repetition rate provided by the fs-CARS system can directly measure the corresponding parameters in the high-speed transient combustion field without relying on conventional time-averaged statistical analysis. The use of detection pulses with a duration of picoseconds (ps) to detect Raman coherence constitutes a femtosecond/picosecond (fs/ps) hybrid CARS system. Using femtosecond/picosecond (fs/ps) purely rotating CARS system to measure combustion plume temperature and O2 /N2 of solid propellant containing aluminum particles under normal pressure The measurement was carried out, and Figure 5 is the schematic diagram of the CARS system used. The difference from propellants that do not contain metal components is that the combustion of metal-based propellants has strong background brightness and scattering of hot metal particles with a diameter of 102 µm, which constitutes an unfavorable environment for laser diagnosis. Traditional CARS or even nanosecond-level CARS pulse lasers are used in aluminum Particle combustion diagnosis is seriously affected by non-resonant background, and it has been confirmed that laser-induced breakdown plasma is generated in the process. The introduction of femtosecond/picosecond (fs/ps) laser pulses improves CARS detection by providing time gating to eliminate strong non -resonant background interference. The background-free spectrum obtained by delayed detection makes the temperature and relative oxygen content fitting values closer. This further proves the strong adaptability of the integrated fs/ps hybrid CARS system in detecting multi-phase environments and combustion of solid propellants containing large particles. fs/ Another obvious advantage of ps CARS is the non-collision nature of the measurement resulting from the ultra-short time scale of laser-matter interaction, within 300~ The measurement error within the 2400K temperature range is approximately ±8%. Improving the optical resolution of the detection system, extending the detection time, or taking other means can effectively enhance its low-temperature sensitivity.

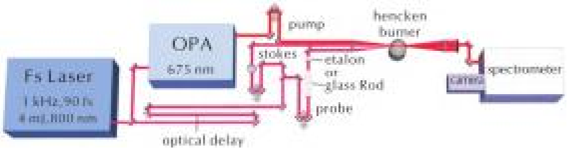


Figure 5 fs/ps CARS temperature measurement system schematic diagram

To sum up, the current application of CARS technology in the field of energetic materials at home and abroad has developed from nanosecond CARS to fs/ps CARS. The reduction of laser pulses by several orders of magnitude allows it to overcome the traditional CARS technology, which is greatly affected by the collision effect. At the same time, high -frequency CARS provides more data in the same time, allowing CARS technology to accurately obtain rapidly changing combustion field information. For a series of problems such as the complex combustion environment and particle scattering of solid propellants containing metal materials, fs/ps CARS technology also provides suitable solutions. Compared with PLIF, a big drawback of CARS technology is that its single-point measurement requires simultaneous spatial and temporal alignment of multiple laser beams and spectral modeling, which may sometimes be difficult to achieve in the complex combustion environment of solid propellant applications, and is accompanied by low spatial resolution. In addition, this technique is difficult to perform at high pressures due to the narrowing of the collision spectral lines.

3 OPTICAL EMISSION AND ABSORPTION

Optical emission and absorption are similar to the light scattering method and also belong to the category of spectroscopic analysis. It has the characteristics of non-contact, fast response, and can measure high temperature and component concentration. However, the disadvantage is that it can only measure thermal equilibrium and no self-absorption. The combustion zone is sometimes difficult to measure point by point, and the error is large. Optical emission and absorption include radiation, RS-FTIR and LIBS dominated emission spectra and LAS dominated absorption spectra.

3.1 Radiation Method

The radiation temperature measurement method is a non-contact passive temperature measurement method. This method selects an appropriate detection period based on the radiation characteristics of the flame source itself to obtain the radiation information of the corresponding band of the field to be measured for temperature measurement. According to Planck's law, researchers have proposed flame temperature calculation methods such as radiation intensity method, single-wavelength method, dual-wavelength method, and multi-wavelength method. Since the emissivity of complex combustion flames is a function of wavelength, in order to eliminate the influence of changes in emissivity with wavelength and improve measurement accuracy, the dual-wavelength method and the multi-wavelength method are more suitable for application in energetic material combustion and solid rocket ramjet engine combustion flow field measurements. Mild.

Yang et al. used radiation spectroscopy to carry out online measurement of the combustion temperature of rocket solid propellant in the harsh environment of the engine and proposed its calibration method and spectral characteristics. Based on Planck's law and spectral fitting method, the corresponding solid propellant combustion was obtained temperature. In Rocketbasedcombined loop (Rocketbasedcombined c ycle, RBCC) engine ground test, an online measurement system using a fiber spectrometer was used to obtain the relationship between the radiation spectrum from 200 to 1100nm and combustion efficiency. This method can effectively diagnose the quality of combustion through combustion temperature parameters and radiation rate parameters. That is, when the radiation rate in the 200~1100nm measurement band is close to 0 and relatively stable, it can be determined that the flame is in efficient and steady state combustion. In the combustion of composite solid propellant, a large number of solid metal particles burn and emit strong light, which causes great noise interference to laser diagnostic technology. However, there is still a radiation signal with a sufficient signal-to-noise ratio during the combustion process, so the radiation method can obtain Better temperature test results.

A dual-wavelength solid rocket motor plume temperature testing method based on flame radiation spectrum was proposed. A test system was built using a 350-1000nm band optical fiber spectrometer. The solid rocket motor plume radiation spectrum measurements of propellant formulas with different aluminum contents were carried out. Through The experimental data analyzed the effects of different aluminum content formulas on the radiation spectrum, plume temperature and emissivity, and provided experimental data support to guide the formula design of aluminum-containing solid propellants. The dual-wavelength method improves its time resolution and measurement accuracy by measuring the spectral radiation of two bands on the same optical path and using the ratio to eliminate interference from external factors such as optical path loss and vibration. However, this method requires that the wavelengths are close and The spectrum is single. In actual engineering measurements, it is difficult to achieve a single spectrum due to spectral diffusion, causing the dual-wavelength method to introduce errors. In order to avoid such problems, multi-wavelength temperature measurement methods that perform ratio processing at the same wavelength have been developed and applied. Pingli used the multi-wavelength radiation temperature measurement system shown in Figure 6 to study the radiation spectrum characteristics of aluminum particle combustion. A dual-machine simultaneous measurement method combining blue light backlight and radiation method was used to measure the combustion process of solid propellant aluminum particles, and analyzed the aluminum particles. The agglomeration process at the burning surface of solid propellants. In addition, three-dimensional temperature field reconstruction can also be performed through radiation temperature measurement and imaging methods. A dynamic combustion field three-dimensional radiation thermometry method based on multi-CCD synchronous coupling was used to measure the temperature of a certain model of fixed tail flame, and the three-dimensional temperature field of the tail flame was inverted. The error was within 8%, and the upper limit of temperature measurement could reach 2000~3000K. Dual-wavelength and multi-wavelength temperature measurement methods have shown good application prospects in real-time dynamic measurement of combustion temperatures of solid propellants, especially high-energy propellants in harsh combustion environments.

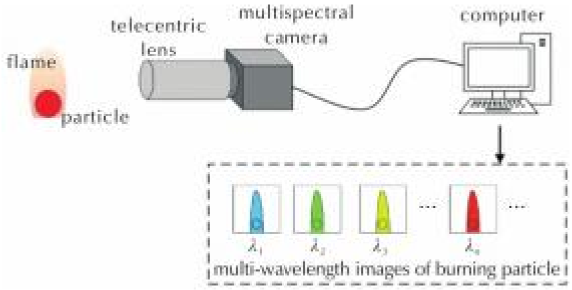


Figure 6 Principle diagram of multi-wavelength radiation method temperature measurement

Compared with other non-contact active temperature measurement methods, the radiation method does not require emitting signals around the flame, making the measurement system relatively simple and easy to use. And because this method only obtains the flame temperature by measuring flame self-radiation, it provides direct heat transfer information and has good measurement stability in strong vibration environments. Even though the application of radiation method in energetic material combustion or engine combustion diagnosis is subject to measurement errors caused by smoke and combustion particles in the test environment, and the measurement accuracy is slightly lower, this method can simultaneously ensure spatiotemporal resolution and can still achieve high accuracy in extreme environments. It can achieve better results and is still one of the important methods for measuring the combustion temperature of aircraft tail flames and energetic materials.

3.2 Laser Absorption Spectroscopy (LAS)

Laser absorption spectroscopy is an absorption spectroscopy technology that uses laser as a light source. Absorption spectroscopy is a macroscopic manifestation of the material absorbing light and transitioning from a low energy level to a high energy level. Materials can be analyzed through the quantitative relationship between matter and light absorption described by molecular spectroscopy and Beer-Lambert's law. Microscopic components and concentration information. Because the laser is highly monochromatic and directional, it has good selectivity for the spectral lines of the measurement object, and because the laser is tunable, it also avoids the added structural complexity of optical devices such as gratings and prisms.

Different types of lasers are available for this diagnostic technique, the most widely used currently being Tunable Diode Laser Absorption Spectroscopy (TDLAS) using tunable diode lasers. TDLAS is a highly sensitive, non-contact combustion diagnosis method based on molecular absorption spectroscopy (the principle is shown in Figure 7). The controller changes the temperature and current to rapidly modulate the semiconductor laser, so that the laser reaches the absorption frequency domain of the target component, and realizes the absorption spectrum Quickly scan and use Beer-Lambert's law to obtain the relevant parameters of the area to be measured by calculating the incident and projected light intensities.

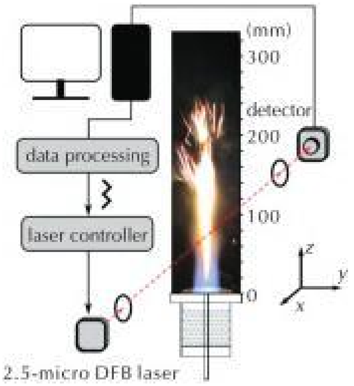


Figure 7 Principle diagram of aluminum particle jet flame TDLAS diagnostic system

Since the wavelength range of the semiconductor laser covers H2 O, CO2, O2 The molecular absorption spectra of combustion products such as these are often used as probes to study the entire combustion process. Use 150 W Pulsed Arc Lamp vs. RDX The propellant was subjected to absorption spectrometry and the important combustion intermediate product CN was obtained. and NH component concentration. The absorption spectrum in the 307 ~ 311nm region was used to measure the flame temperature and OH concentration of the double-base propellant under a high pressure of 69.1atm. Although the flame temperature obtained at high pressure was in good agreement with the chemical equilibrium calculation, the measured OH concentration was lower than the calculated value. 40%, a difference the researchers attribute to the high sensitivity of OH concentration to local flame stoichiometry and temperature changes.

In recent years, TDLAS has been applied to high-pressure combustion flow field diagnosis in engines, which can provide detection of combustion process status, intermediate products, airflow velocity and other information for the combustion flow field of aviation, rocket and other engine combustion chambers. The TDLAS technology was used to measure the water vapor concentration and temperature at the combustion chamber outlet of the scramjet engine. The standard deviations between the measured values of temperature and water vapor concentration and the actual values were 4 respectively. 2% and 2.7%, which are generally similar to the CFD simulation results. The modulation parameters were used to calculate calibration constants, and the TDLAS second harmonic method was used to measure the transient supersonic flow field of the supersonic nozzle. The measured average temperature of the combustion chamber exceeded 1832K, with a standard deviation of 53K. After analysis, it was found that the source of the measurement error is the flame oscillation in the combustion process, and the system error is the deviation between the theoretical and actual values of the spectral line parameters.

In terms of propellant combustion, utilizing TDLAS Wavelength modulation spectroscopy technology measured the combustion flame zone temperature and CO concentration of AP/HTPB composite propellants containing aluminum particles of different particle sizes and without aluminum using mid-infrared wavelengths, while AlO emission spectroscopy technology was used to measure the temperature of the aluminum particle combustion zone. Experimental results show that on the basis of equal time, micron-sized aluminum particles burn in a diffusion-controlled state, while nano-aluminum particles burn in a kinetically controlled combustion state or close to a kinetically controlled combustion state. Subsequently, the gas temperature and HCl concentration in the flame during the combustion of metal -containing AP/HTPB composite propellant were studied. The results showed that the combustion temperature of aluminum-lithium propellant was 80~200K higher than that of aluminum propellant, and the combustion was more complete. It also confirmed the application of TDLAS technology. Effectiveness in measuring propellant combustion flame temperature and combustion product concentration. Research has found that combining TDLAS technology with the Doppler effect can measure the airflow velocity in a supersonic flow field. Select the absorption spectrum line of H_2O at the center wavelength of 1391.7nm, and use TDLAS The direct absorption spectroscopy method was used to conduct static and dynamic measurements of the rocket skid solid propellant charge combustion plume velocity. The average flow velocity during the entire ignition process under static conditions was measured to be 1057.5m/s, and the average flow velocity under dynamic sliding conditions was 1249.8m/s, which confirmed the Applicability of TDLAS system in static and dynamic measurements of solid propellant charge combustion plume velocity.

In addition to TDLAS technology, the LAS-based method uses an ultrafast laser absorption spectroscopy (ULAS) diagnostic method to measure and characterize the combustion flame temperature and temperature of AP/HTPB and AP/HTPB/Al propellants under different pressures (1~40bar) at kHz. CO concentration. The results show that mid-infrared ULAS technology can provide high-fidelity, calibration-free temperature and gas concentration measurements with sub-nanosecond time resolution in a typical rocket engine high-pressure combustion environment.

For testing the flow field of aviation and rocket engine combustion chambers and the combustion flame of energetic materials, the optical absorption method has the same advantages as the light scattering method, which are non-contact and fast time response, and is suitable for measuring small molecules and free radicals in the combustion flame. and changes in the combustion flow field. However, for flames with large temperature gradients, optical measurement errors are often caused, and the resolution is lower than other optical methods; for macromolecules and their component concentrations, because it is difficult to measure point by point, the spectral peaks of macromolecules overlap at high temperatures. and other factors lead to inability to accurately measure. However, its advantage over technical methods such as CARS is that its cost is not high, and it can be flexibly applied to various combustion environments without complex operations. Based on the shared optical port, it can be combined with optical methods such as high-speed photography and radiation temperature measurement. achieve the purpose of measurement.

twenty three Remote sensing Fourier transform infrared spectroscopy (RS-FTIR)

Remote sensing Fourier transform infrared spectroscopy (RS-FT-IR) is a typical infrared emission spectrum testing method. Its principle is based on the Pin the fine structure of the rotating baseband established by Herget et al. or R-branch spectroscopy method of measuring the temperature of hot gases, based on absorption measurements along the atmospheric path between a radiation source and a spectrometer. Its advantage is that the path can be extended to tens of meters to several kilometers, and the temperature and product component concentration can be measured over long distances in dangerous combustion environments. RS-FTIR technology has been applied to the combustion temperature of energetic materials. Measurement.

A passive remote sensing Fourier transform infrared spectrometer was used to measure the combustion flame temperature of solid propellants mixed with nanoscale metal oxides, mixed with common metal oxides of the same material, and without adulterants. FTIR The instrument resolution is 1cm -1. The combustion flame temperature is calculated based on the baseband emission spectrum structure of the H_2O molecule in the combustion product and the molecular vibration spectrum thermometry. The results show that doped with nanoscale CuO, Fe_2O_3 and NiO particles The combustion flame temperatures of the solid propellant are 3089 and 3193 respectively. and 3183K. This temperature is not significantly different from the combustion flame temperature of ordinary metal oxides mixed with the same material and solid propellant without adulteration. The RS-FTIR technology was used to measure the radiant brightness of the solid propellant combustion flame, and the curves of the combustion integrated brightness (radiant energy) of the two propellants changing with time were obtained. propellant gas composition and combustion characteristics information. Using a RS-FTIR spectrometer combined with a telescope, the infrared emission of nitroguanidine/AP/PTFE solid propellant was collected from 30m away, and a spectrum between 800 and 4700cm-1 was obtained. Its different combustion was determined through the HF and HCl anti-vibration bands. Temperature of time, spectral resolution 2cm-1.

The results show that RS-FTIR emission spectroscopy is a fast and sensitive method for testing flame temperature and product concentration, which can be used to diagnose the combustion process of energetic materials.

Although RS-FTIR technology has been proven to be able to better measure the combustion temperature of solid propellant and the influence of components on combustion temperature in a wide band range, it has high signal-to-noise ratio, high sensitivity, fast detection speed, and can identify the emission source. Absolute energy spectrum and other advantages, but commercial FT-IR instruments are expensive and have limited time resolution. The measurement results are sight range and path averages and are difficult to quantify. Factors such as high-pressure conditions accompanied by unstable combustion are still the mainstay of RS-FTIR technology in solid propellants. Problems faced in combustion diagnosis.

twenty four Laser Induced Breakdown Spectroscopy (LIBS)

Laser-induced breakdown spectroscopy (LIBS) technology is an advanced detection method based on atomic emission spectrometry analysis technology. It is extremely useful in the fields of qualitative and quantitative measurement of intermediate product elements in various complex combustion flow fields such as solid, liquid, and gas combustion flames. Development potential. The basic principle of LIBS technology is to use a high-energy pulsed laser to focus on the sample surface and interact with the sample. The atoms and molecules in the laser focus area absorb energy to generate laser plasma. The ion begins to cool immediately after the laser pulse terminates, and is in an excited state during the process. Atoms and ions jump from high energy levels to low energy levels to release light with specific frequencies, that is, the characteristic spectral lines of the corresponding atoms and ions. The component elements of the analyte can be obtained by detecting the wavelength of the characteristic spectral line, and the calibrated intensity of the characteristic spectral line can reflect the concentration of the target element in the analyte and other related information (Figure 8). Because the laser pulse energy is high, it can penetrate deep into the flame, the pulse time is short, and the range of action is small. Related research has been devoted to the application of this technology in combustion diagnosis. Figure 8 is a schematic diagram of the LIBS combustion diagnosis system.

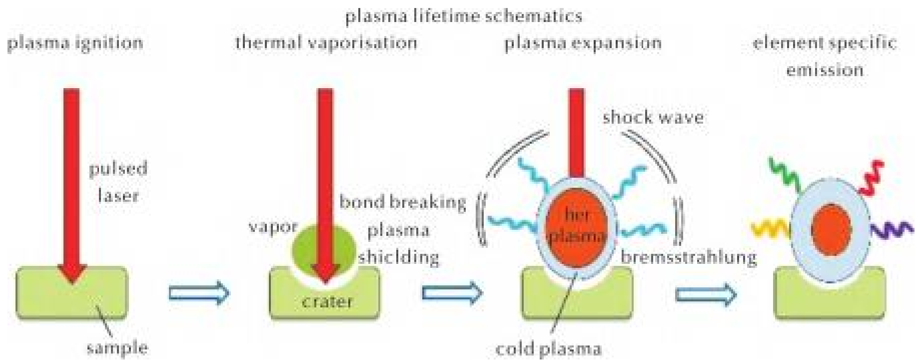


Figure 8 Schematic diagram of laser-induced breakdown spectroscopy

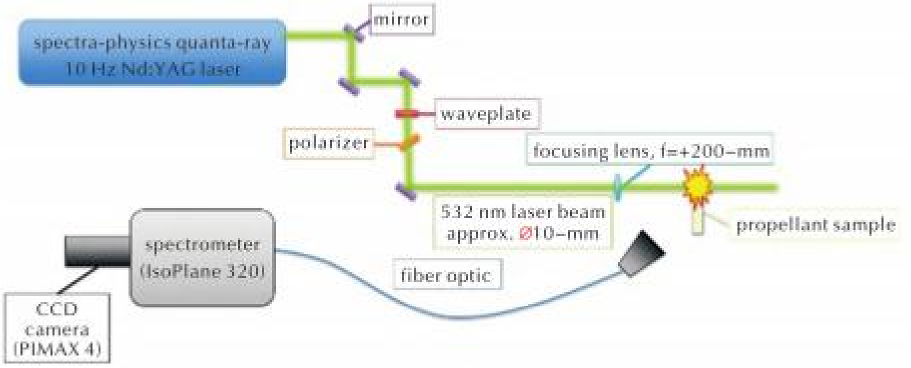


Figure 8 Schematic diagram of LIBS combustion diagnostic system

The most typical application of LIBS technology in the field of combustion is the quantitative measurement of flame equivalence ratio (Φ) to reflect the impact on the reaction process and the ratio of oxidizer and fuel to combustion products. The H/N peak intensity ratio and fuel equivalence ratio were calibrated, and a calibration curve of the H (656nm) spectral line half-height width and temperature was established for combustion jet flame detection. LIBS was used to detect the local carbon-to-hydrogen ratio in the ethylene partially premixed flame, and the relationship between the gas flow O/C peak intensity ratio and the concentration equivalence ratio was pre-calibrated, and then the distribution of carbon and oxygen atomic ratios in the flame axial height under different equivalence ratios was studied., which is consistent with the simulation results, confirming the effectiveness of LIBS technology in the combustion flow field.

In recent years, researchers have applied LIBS technology to the defense sector to characterize the release of heavy metal elements when energetic materials burn. In 2017, he first used a 10Hz solid laser as the excitation source to study the characteristic spectral lines of metal elements in the combustion flame of solid propellant containing trace amounts of metal. LIBS characteristic spectral lines were detected in solid propellant combustion plumes doped with known concentrations of aluminum, and the optimal energy for generating plasma was determined through studies of laser energy correlation, resulting in detection with a higher signal-to-noise ratio.. In order to reduce the impact of laser-plasma interaction and background noise interference on the experimental results, the above experiment was repeated using an ultrashort pulse laser with a pulse duration of 80fs at 1kHz (Figure 9 shows the peak spectrum of the solid propellant with an Al mass fraction of 16% Image), the result is a better linear relationship between the LIBS emission line intensity and the Al concentration initially present in the line beam (see Figure 10), confirming the superiority of ultra-short pulse LIBS technology and its application in aluminum-containing propellants Applicability in combustion diagnostics. Using a 10Hz Nd:YAG laser, the concentration of gas phase iridium was measured using LIBS technology for the first time. Iridium is one of the most commonly used catalysts in single propellants, and its damage and consumption during propellant combustion is one of the main factors affecting the life of single propellant. The significance of this work is to make LIBS technology have an important role in propulsion. Prospects for active monitoring of single propellant health by detecting propellant plumes during propellant combustion.

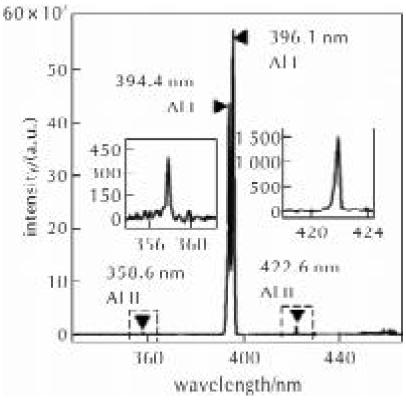


Figure 9 LIBS spectrum of solid propellant with 16% Al mass fraction

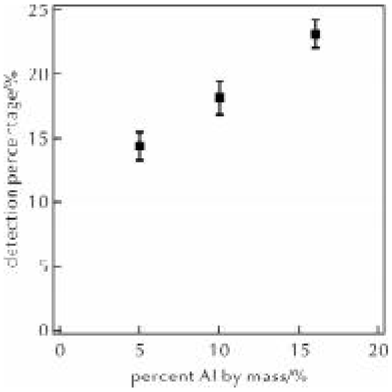


Figure 10 Solid propellant signals with Al mass fractions of 5%, 10% and 16%

The relationship between intensity and mass concentration LIBS is widely used in in-situ rapid measurement of multiple types of products. It has high accuracy in measuring the gas fuel equivalence ratio and the concentration of metal atoms in solid fuel flames. It can also obtain non-metallic materials such as C, O, H, and N. Characteristic spectral lines of metal elements enable measurement of multi-product components and multi-characteristics. At the same time, LIBS has gradually developed combustion diagnosis near the wall of the liquid rocket combustion chamber to determine the relative concentration of the main combustion substances and reduce the uncertainty of CFD verification. It has only been developed and utilized in the field of energetic material combustion in recent years, mainly focusing on the quantitative detection of metal elements. The disadvantage of LIBS technology in this field is that it may be affected by factors such as the stability of the plasma signal of suspended particles in the gas, high-energy laser intrusion, and high pressure in the combustion chamber, resulting in large signal fluctuations.

4 IMAGING METHOD

This type of method uses optical imaging principles such as interference to capture and image the results, often combined with high-speed photography and holography (DIH) technology, to measure the combustion flow field velocity and density gradient, and reflect the flame structure and changes over time. Compared with In terms of light scattering and optical emission and absorption in the spectral range, imaging methods are more biased toward macroscopic measurements.

4.1 Particle Imaging Velocimetry (PIV)

Particle imaging velocimetry (PIV) is a testing technology based on elastic scattering technology for instantaneous velocity spatial distribution, which can conduct plane and spatial testing of combustion flow fields. The basic principle of PIV is to continuously use laser pulses to take two spatial distribution images of fluid particles in a very short time. The images show the displacement size and direction of the flowing particles. The particle velocity vector is calculated through correlation analysis. Figure 11 is a schematic diagram of the PIV system.. Compared with technologies such as single-point laser Doppler velocimetry (LDV) and phase Doppler particle analysis (PDPA), PIV is more inclined to test and study the overall flow field, and has been widely used in various engine combustion flow fields and liquid fuels. In the study of spray particle motion.

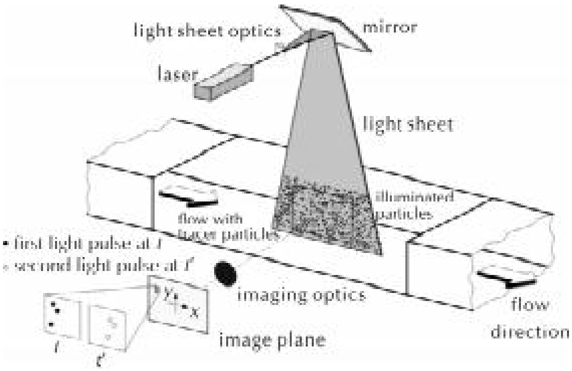
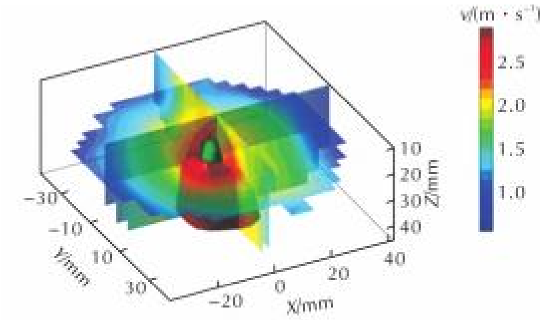


Figure 11 Schematic diagram of particle imaging velocimetry system

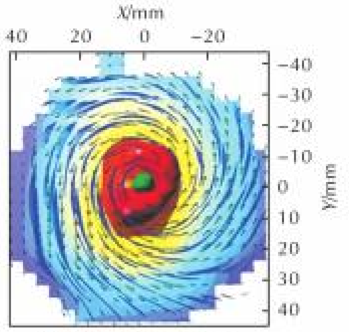
PIV velocity measurements were carried out in the exhaust plume of a small solid rocket motor. The plume velocity in this environment reached 630m/s. Although the temperature exceeded 3000K and the heterogeneous particle distribution inside the plume caused a large amount of vector loss, the experimental results demonstrated it well. The applicability of PIV using jet particles as tracer particles in plume measurements was demonstrated. The small size of rocket engines makes optical access with shorter optical paths easier to achieve, however observing solid propellant plumes in larger rocket engines suffers from increased optical path lengths and reduced transparency due to increased particle scattering, and in larger devices The application of propellant combustion diagnostics is still the development direction of PIV technology. PIV was used to measure the flame flow field velocity under different total pressure losses in the cold state of the aeroengine recirculation combustor. MgO with an average particle size of 10 μm was used as a tracer particle to fully mix with the airflow, and the pressure loss and combustion velocity in the recirculation combustor were obtained. Dynamic relationship. Using silicone oil particles with an average size of 1μm as tracer particles, PIV technology was used to observe the engine speed of 3500r/ The velocity field distribution at the center of the combustion chamber in the working cycle at min. In high-temperature and high-pressure environments, since PIV technology uses Mie scattering to be insensitive to high-pressure environments, and strong background light can be filtered by adding a filter, the test results are less disturbed and more accurate.

In recent years, PIV technology is developing to a multi-dimensional level in engine combustion flow field diagnosis. First of all, stereo PIV technology was developed, a single camera was replaced with a dual-camera group, and the test results of the three velocity components in the two-dimensional space of the combustion flow field were obtained, allowing researchers to have a more intuitive understanding of the spatial flow conditions of the complex flow field. Secondly, a tomographic PIV technology was developed, using 4 CCD cameras to simultaneously shoot the flow field at different spatial positions. Through the two-dimensional projection of the tracer particles, the tomographic algorithm was used to reconstruct the three-dimensional structure, thereby realizing the engine combustion chamber. Transient measurement of three velocity components in three dimensions near the tumble plane. In the body PIV part, a double-pulse YAG laser is used as the light source, and four CCD cameras are positioned in the vertical direction of laser irradiation to take pictures. Through high-resolution images (Figure 12 shows the PIV reconstruction results of the swirling flame body), the temperature and velocity coupling effect on the combustion flow field is obtained. The resulting influence patterns verify the feasibility of this quantitative, multi-dimensional, and visual comprehensive optical technology in diagnosing complex flame flow fields. This three-dimensional velocity vector field measurement technology (Tomo-PIV), which combines planar PIV and tomographic reconstruction algorithms, is a current research hotspot in the field of three-dimensional flow field velocity measurement. This method is suitable for high-density tracer particles, does not require coherent light illumination, and obtains flow The field spatial resolution is higher and the imaging quality is better, and it is expected to be widely used in three-dimensional velocity measurement research on complex turbulent flow fields of energetic materials and aerospace engines.

In terms of research on the combustion of energetic materials, we combined PIV, high-speed photography, FTIR and other technologies to conduct visual research on the flame flow field and temperature field of pyrotechnic powder combustion. We used PIV technology to observe the gas-particle two-phase flow in the flame flow field, and analyzed The combustion reaction mechanism of this composite energetic material. In the study of muzzle flow field velocity measurement, in order to verify the applicability of PIV technology in this extremely challenging supersonic environment, comparative experiments with PIV, high-speed schlieren, optical particle counters, combustion chamber pressure sensors and other technologies were used. 300BLK ammunition propellant combustion flow rate. The test results show that PIV technology can accurately distinguish the main characteristics of the flow and the instantaneous velocity field. The sub-micron particles in the combustion products can follow the gas flow with high fidelity and serve as tracer particles. The solid ZrO2 coated on the propellant The pellets also have good flow fidelity and are suitable for combustion in subsonic conditions.



(a) Velocity cloud maps and isokinetic surfaces of different cross-sections



(b) Streamline diagram

Figure 12 PIV reconstruction results of swirling flame body

While PIV technology is developing towards high dimensions and high spatiotemporal resolution, the comprehensive diagnostic method combined with PLIF or PDPA, schlieren and other technologies further enriches the visual information of the combustion flow field and is useful in diagnosing complex combustion processes of energetic materials and engines. It provides more comprehensive image information in complex combustion environments and has great development potential.

4.2 Schlieren Method

Schlieren technology is a flow field display technology that was first proposed by Topler in the 1880s. The schlieren method uses the principle that when a beam passes through a flow field with a density gradient, the beam will be deflected due to a change in the refractive index, and then the deflected beam is converged, and a knife-edge shield is used to form a stripe image after the beam diverges, and the light and dark areas of the image are Represents the density change of the flow field. Typical light paths of the schlieren method include Z-shaped and T-shaped light paths. The Z-shaped light path is the most commonly used light path for jet research, while the T-shaped light path is mostly used in burners such as engines with a single window. According to different combinations of light sources and holes, the schlieren method can also be divided into ordinary schlieren and laser schlieren. In recent years, with the help of high-frequency lasers and flash lamps, or combined with high-speed photography, the time resolution and brightness display of the schlieren method have been greatly improved, making it widely used in engine combustion flow fields and energetic material combustion diagnosis.

Combining the ignition delay and combustion characteristics of paraffin-based fuels with solid amine borane, chemiluminescence and high-speed schlieren imaging techniques were used to identify different steps of the combustion process. The reduction in ignition delay of the viscosity-modified rocket propellant was clearly observed, confirming that the shorter There is a clear correlation between ignition delay and higher polymerized α-olefin concentration, which provides an effective research method for studying the comprehensive impact of other condition changes such as subsequent pressurization and addition of rheological additives on ignition delay. Focused schlieren technology was used to study the aluminum agglomeration mechanism on the burning surface of HTPB/AP/Al propellant. The flame structure information during propellant combustion was obtained (Figure 13), and the melting, ignition and combustion during the evolution of the agglomerates were analyzed. and other microscopic processes, and statistics of parameters such as particle size distribution and velocity were used to verify the numerical simulation results of propellant combustion.

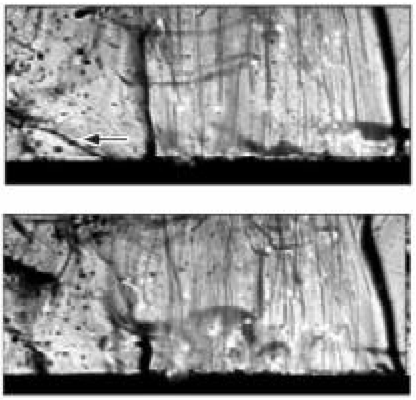


Figure 13 HTPB/AP/Al propellant flame propagation image captured by focusing on schlieren technology

Research results show that schlieren technology can effectively display the structure and development process of solid propellant flames, and can also track and monitor the alumina particles generated by combustion. A new algorithm based on focused schlieren technology for solid propellant containing inert particles to burn surface particles and aggregates is proposed. Compared with the study of the aluminum agglomeration phenomenon on the burning surface of propellant, this method focuses more on detecting the protrusions of continuous one-dimensional curves. About 84% of the particle size detection of a certain type of propellant is close to the marked particle size, with an allowable deviation of 25%. When applied During the combustion of aluminum-containing solid propellants, particles and aggregates can be detected before they begin to coalesce. The newly developed background-oriented schlieren technology (BOS) is the current research hotspot in schlieren imaging. BOS Correlation calculations applicable to the schlieren method can be performed on the image background. Although it is different in principle from the schlieren effect, it can still image the density gradient and obtain an image effect similar to the schlieren, so it is also used in sprays. Research.

Schlieren technology does not require complex optical paths and expensive laser light sources, the application conditions are relatively simple, and the measurement cost is relatively low. When studying the combustion of energetic materials, the thin combustion zone under high pressure reduces the resolution of the ordinary schlieren method. The reflection from the observation window may also interfere with the schlieren image. Similar to the direct optical photography method, there is a single focal plane and excessive depth of field. Narrow and other shortcomings reduce measurement efficiency. The use of laser monochromaticity and bandpass filters can largely solve the problem of flame self-illumination and interference, combined with high-speed photography and digital coaxial holography (DIH), it is also possible to obtain instantaneous surface schlieren patterns of energetic materials suitable for the working conditions of the schlieren method in combustion experiments.

5 CONCLUSION

Optical diagnostic methods play an important role in the experimental diagnosis of energetic material combustion. With their advantages of non-contact, no interference, and real-time monitoring, they have become a powerful tool for contemporary research on engine combustion flow fields and energetic material combustion phenomena. For engine flow, Provide guidance on field design and energetic material formulations and combustion mechanisms. As far as current research is concerned, optical diagnostic methods have been widely used in combustion diagnosis of energetic materials. However, due to the relatively harsh combustion environment of energetic materials and engines, different optical methods have their limitations and applicability. A single test There are certain deficiencies in the experimental parameters available through technology. Research should combine different diagnostic methods based on actual conditions and needs, and use their respective complementary advantages to achieve more accurate experimental research on multiple parameters of the combustion process. Based on the above issues, this article reviews the application of several optical diagnostic methods in the study of energetic material combustion and engine combustion chambers in recent years. It summarizes the measurement principles and objects of various methods, analyzes their respective advantages and disadvantages, and provides guidance for the future. Optical diagnostic methods for studying the combustion of energetic materials provide reference. The specific conclusions are as follows:

(1) Methods such as radiation method, LIF and LAS have great application potential in carrying out multi-dimensional temperature field measurement research. They can be widely used in measurement research on energetic material combustion and aerospace engine combustion chamber temperature fields, and further develop three-dimensional temperature field reconstruction, etc. Work.

(2) PLIF technology has superiority in testing combustion intermediate products and is sensitive to detecting concentration changes of free radicals and molecules such as OH, CH, CN, NO, HCO, CH2 O, etc., as well as metal atoms such as Al and Fe during the combustion process. The above groups and atoms are also important tracer products

that characterize the flame structure of energetic materials. By studying the concentration distribution and changes of intermediate products, the reaction kinetic model of energetic materials can be further improved and verified, and microscopic data can be provided for the combustion reaction mechanism. support.

(3) Technologies such as radiation method, LAS and LIBS have development potential in terms of equipment miniaturization and portability, and LAS has a shareable optical port, which can combine radiation method and optical methods such as high-speed photography for simultaneous measurement. The optimization and improvement can carry out synchronous online diagnosis of the temperature field and flow field structure of the engine combustion chamber under more environmental conditions.

(4) Schlieren method and PIV have great development potential in multi-dimensional flow field display and multi-velocity component measurement research. They can be more applied in the testing of aviation engine combustion flow fields. By combining direct shooting with high-speed cameras and PLIF and other optical methods to build a more suitable and efficient optical diagnosis system to obtain more microscopic and rich multi-dimensional data information.

COMPETING INTERESTS

The authors have no relevant financial or non-financial interests to disclose.

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