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Single-pulse CARS spectra in solid propellant combustion at atmosphere pressure

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Received March 19, 2003

The unstable-resonator spatially enhanced detection (USED) coherent anti-stokes Raman spectroscopy (CARS) measurements of temperature and N₂ concentration in the combustion of solid propellant at atmosphere pressure are reported. The USED CARS measurement system has a high spatial solution of ~ 0.1 mm in diameter and 3 mm in length, and permits instantaneous measurement at 10-Hz rate. The single-pulse N₂ Q-branch CARS spectra have been obtained from the propellant combustion. The temperatures and N₂ concentrations of the propellant flame at different height have been achieved by fitting the experimental data to theoretical spectra. The results indicate that the temperature is up to ~ 2500 K with N₂ concentration in a range from 10% to 26%.

OCIS codes: 300.6230, 120.1740, 120.6780, 120.4820.

Knowledge of combustion species and temperature is necessary for accurate modeling of solid propellant reaction mechanisms. Diagnostics of solid propellant combustion are extremely difficult because of the high luminosity of particle laden, turbulent flame, and small burning volume. The conventional temperature measurements including thermocouples may perturb the flames, require extensive interpretation to yield valid results, and provide no information about the molecular constituents. Among the various laser-based diagnostic techniques, laser-induced fluorescence (LIF) has been used to measure temperature and trace species [1], such as OH and NO, but analysis of LIF data is difficult because of the effects of collisional quenching. Vibrational Raman scattering (VRS) is capable of detecting temperature and major species in CH₄-air flames [2], but VRS is only suited to a "clean" environment with little interfering emissions because VRS suffers from its inherent weakness. For example VRS is roughly a factor of 1000 weaker than Rayleigh scattering (for N₂). It has been demonstrated that coherent anti-stokes Raman spectroscopy (CARS) is a powerful and non-intrusive diagnostic method for determining the temperature and species concentrations in a number of practical combustion devices [3–6]. CARS is well suited for hostile environment diagnostics because of its coherence, high conversion efficiency, spatial and temporal precision. Unstable-resonator spatially enhanced detection (USED) CARS is a two-beams 3-D phase-matching configuration [6] which can possess high spatial resolution without the alignment difficulties of multiple beams pumping, and the technique has been widely used in areas such as internal combustion (IC) engine [4] and supersonic combustion [6]. In this paper, the USED CARS technique is applied to solid propellant combustion at atmosphere pressure. Measurements of temperature and N₂ concentration from experimental CARS spectra are presented.

A schematic diagram of the optical layout is shown in Fig. 1. A frequency-doubled Nd:YAG laser operating at 10 Hz produces light of 532 nm with a typical pulse width of 8 ns. The most energy of the light beam from Nd:YAG laser is used to pump the broad-band dye laser employing Rhodamine640 to provide the stokes beam (ω₂) for N₂ Raman resonance with full width at half-maximum of ~ 130 cm⁻¹, and the rest energy is used as pump beam (ω₁) for the CARS process. Optical routings are designed to ensure that the 8-ns laser pulses are temporally synchronized when they arrive at the field lens. The pump beam is reflected by a mirror (M₃) with a 5-mm aperture and forms an annular pump beam satisfying USED CARS phase-matching configuration. The Stokes beam, after passing through the 5-mm aperture in the mirror (M₄), follows the axis of the pump beam and is positioned in the center of the annulus without overlapping the pump beam. The CARS signal beam (ω₃ = 2ω₁ − ω₂) generated in the focal region of ~ 0.1-mm diameter by 3 mm long is collimated by the field lens in the receiver. The ω₁ and ω₂ beams are split off by a dichroic mirror, and the CARS beam, after additional spatial and spectral filtering, is focused into a fiber-optic cable. The fiber-optic transports the signal to the spectrographic instrument. Single-pulse spectra are acquired at the laser repetition frequency and subsequently stored in a computer for analysis. The accuracy of single-shot

![Fig. 1. The USED CARS experimental setup.](http://www.col.org.cn)
CARS measurements have been performed in premix H₂-air flames before CARS diagnostics of solid propellant combustion. The uncertainty of temperature measurement is less than 4% for T = 1700 K, and the uncertainty of N₂ concentration measurement is less than 5%.

The experiments have been performed in the solid propellant combustion at atmospheric pressure. The 20-mm black strands of the propellant have a 5 × 5 mm² cross section, and its burn lasts ~ 8 s with a burning rate of 2.5 mm/s. The fixed laser beams are initially focused 5 mm above the propellant surface which regresses after being ignited with NiChrome wire. There are about 80 single-shot CARS spectra stored in a computer during the propellant burning. Figure 2(a) is a typical single-shot nitrogen Q-branch CARS spectrum taken from the propellant combustion when the strand has regressed 19.3 mm away from the laser focus. The height above the propellant surface was calculated on the basis of the propellant burning rate and the record time of single-shot CARS spectrum. In single-shot CARS spectra one can see the effect of the random noise fluctuations from the broad-band dye laser. A typical single-pulse dye spectrum generated in a nonresonant reference gas is showed in Fig. 2(b). The fine structure leads to irregularities in the single-pulse CARS spectrum. The distortion arising from the dye spectrum noise is a main source of measurement errors. Fortunately, the general spectral shape of the dye laser is reproducible from pulse to pulse, and the quality of the single-pulse nitrogen CARS spectra is good enough to deduce the temperature and N₂ concentration of the propellant combustion.

The profile of the nitrogen CARS spectrum is sensitive to temperature and nitrogen concentration in the range from 0.5% to 30%. For this low nitrogen concentration satisfying calculation condition, the temperature and nitrogen concentration can be determined by fitting calculated CARS profiles to the experimental spectra [6,7]. Figure 3 is a theoretical spectrum (solid line) fitted to the experimental data (open circles) adopted from Fig. 2. To correct for the intensity profile of the dye laser and for the non-uniformity of the interference filters, the CARS spectra were divided by a time-average nonresonant background spectrum obtained before the CARS temperature measurement. The broadened fundamental band, high-amplitude hot band and the evidence of the second vibrational band indicate quite a high temperature. The theoretical fits represent a temperature of 2500 K and N₂ concentration of 10%. The CARS spectra from the propellant combustion are analyzed, and the results of temperature and N₂ concentration as a function of distance above the propellant surface are showed in Fig. 4. The dots are experimental data, and the solid lines are least-squares fits to the data points. The temperature remains reasonable constant in the region from 12.5 to 19 mm above the surface, and the average value of these data is about 2500 K. The flame starts to cool off after ~ 19 mm away the propellant surface, and the reason may be that the ambient air diffuses into the focal region close to the boundary of the propellant flame. The fitted line of N₂ concentration indicates that it increases linearly from 10% to 26% with the increasing distance from the surface arising from non-equilibrium parameters process of the solid propellant combustion.

The USED CARS technology was successfully applied to diagnose temperature and N₂ concentration in the exhaust of solid propellant burning at atmospheric pressure. The single-pulse N₂ Q-branch CARS spectra were obtained from the propellant combustion. The quality of the single-pulse CARS spectra was high enough to

![Fig. 3. Comparison of theoretical and experimental CARS spectra.](image)

![Fig. 4. Distribution of temperature and N₂ concentration at different height in solid propellant combustion.](image)
deduce the temperature and $N_2$ concentration of the propellant combustion. The CARS data were analyzed by comparison with theoretical spectra, and the temperature and $N_2$ concentration of the propellant combustion were presented as a function of distance above the propellant surface. The measured results indicate that USED CARS is well suited for quantitative diagnostics of solid propellant combustion at atmosphere pressure.

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