Multi-pass Laser Raman Spectroscopy for Combustion Diagnostic

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Summary:

In this paper is proposed a laser-based system to perform a real time Raman spectroscopy applied to in-line combustion diagnostic. This approach is widely used for solid and liquid analysis; the problem related to gas samples is the low density since the amount of Raman scattering is proportional to the quantity of molecules in the matter-light interaction region. Our setup is based on a multi-pass cell (designed to increase the collected signal) that allows high-frequency acquisitions.

Keywords: combustion, diagnostic, Raman, spectroscopy, laser

Background, Motivation and Objective

Solutions to improve combustion efficiency have experienced a considerable growth in the last years. Such analysis can lead to substantial cost reductions, paving the way towards more sustainable policies regarding air pollution. In order to fully understand the dynamic of industrial combustion, new tools and diagnostic techniques need to be developed.

Optical analysis techniques represent an excellent choice as they are capable to provide high data sampling rate and non-specific sample preparation is required. In addition they are non-destructive and contactless analysis. The main optical techniques are IR absorption spectroscopy, such as FT-IR or tunable diode laser (TDLAS) approach, and Raman spectroscopy.

Raman spectroscopy is based on the phenomena of radiation scattering. The sample molecules are excited using a laser source and part of the scattered light will have a wavelength characteristic of the molecule that emitted it. The use of a monochromatic radiation allows to observe different gases with a single source. In addition, as we operate in the visible light range, the use of infrared detectors is not required. The problem related to gas analysis is the low density since the amount of Raman scattering is proportional to the quantity of molecules in the matter-light interaction region.

As demonstrated by these studies [1][2], it is possible to use the Raman technique to effectively study gaseous samples.

Our setup is based on a multi-pass cell (designed to increase the collected signal) that allows high-frequency acquisitions.

Method

Through a dispersion grating spectrometer (coupled with a CMOS camera), we collect the Raman scattering produced inside a windowed cell were a Nd:YAG 532 nm CW laser is focused into the gaseous stream. The interaction cell is coupled to a system of spherical mirrors in order to perform 16 reflections of the laser beam back to the focal plane (i.e.17 passages). This design increases the optical power density in the light-matter interaction region in a way to boost the amount of collected signal.

In Figure 1 is reported the multi-pass setup with its main components: 532 nm CW laser source (1), spherical mirrors (2a, 2b),windowed gas cell (3) and spectrometer (4).

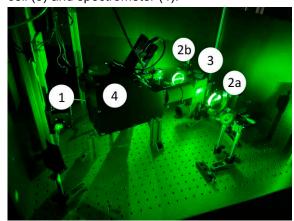


Fig. 1. Multi-pass laser beam setup.

This design is tailored to improve the amount of collected Raman scattered signal in order to minimize the acquisition integration time by maintaining high spectral performance. Figure 2 shows a comparison between the raw spectrum of ambient air collected with the mono-pass and

the multi-pass set up (with the same integration time). A sensitivity increase is clearly visible (appearance of water peak not visible in monopass setup) at the expense of an increase in background diffused light, that has been subtracted using a polynomial 3-rd-order fit.

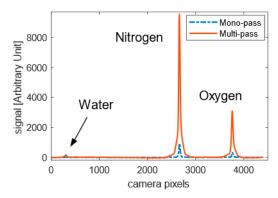


Fig. 2. Ambient air spectrum for the mono-pass system compared with the same for the multi-pass configuration.

The addition of a spherical mirror (feedback mirror) opposed to the spectrometer has also been evaluated. Such implementation reflects the Raman scattering on this side into the spectrometer reaching a further doubling of signal.

Starting from each acquisition frame we compute the spectrum as elaboration of pixel's intensity, then we perform the quantitative analysis using a non-linear least square approach based on the reference gas calibrations.

The application is focused on combustion diagnostic, but as previously mentioned Raman spectroscopy allows to observe the presence of many gases simultaneously. It is therefore possible to extend the analysis to a large group of gases.

Results

In terms of signal intensity, the multi-pass setup reaches a x16 magnification effect compared to the mono-pass system (evaluated on the air Nitrogen integral signal). The presence of the feedback mirror increases the magnification factor up to about x30. These factors are computed from the ambient air analysis without the gas cell. The presence of the cell induces a progressive loss in the laser beam power, in this case we reach a signal magnification of about x14.5 times the single passage (without the presence of the feedback mirror). The system performs analysis with 0.15 seconds per acquisition, which is a significant achievement in Raman spectroscopy on gaseous samples.

An example of a qualitative combustion analysis of a 4-stroke gasoline engine is reported in Figure 2. As we can see the trend of different

gasses is recorded by the system, in this case we focused the analysis on N_2 , H_2O , O_2 , CO, CO_2 and exceeding combustible gas (HC).

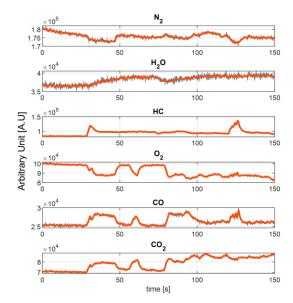


Fig. 3. : Example of qualitative diagnostic of a 4-stroke gasoline engine combustion (0.15 seconds of integration time).

Conclusion

Our research demonstrated that a Raman spectroscopy approach meets the main requirements of combustion diagnostic since it is capable to follow the time scale of the process. Future studies will be focused on further increase in performance. The system will be redesigned in order to get a compact prototype.

Particular attention will be put to increase the reliability of the alignment and to the miniaturization of the system. The goal is to create a portable instrument in order to perform combustion diagnostic in harsh conditions.

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References

- [1] L. Cocola, F. Melison, N. Scarabottolo, G.Tondello, L. Poletto, Diode-based Raman sensor for fuel gas analysis, Proc. SPIE 11354, Optical Sensing and Detection VI, 113541A (2020).
- [2] M. A. Buldakov, V. A. Korolkov, I. I. Matrosov, D. V. Petrov, A. A. Tikhomirov, and B. V. Korolev, "Analyzing natural gas by spontaneous Raman scattering spectroscopy," J. Opt. Technol. 80, 426-430 (2013)