



Full Length Article

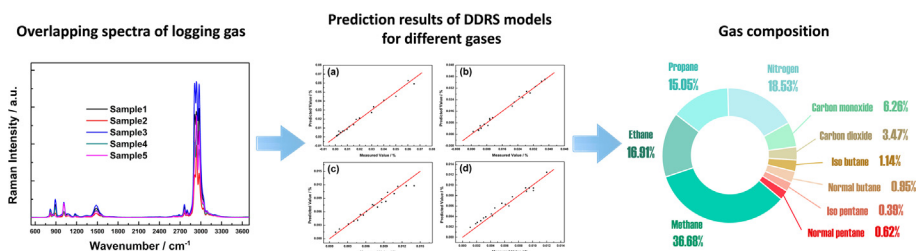
On-line multi-component analysis of gases for mud logging industry using data driven Raman spectroscopy

Xi Han^a, Zhi-xuan Huang^a, Xiao-dong Chen^b, Qi-feng Li^{b,*}, Ke-xin Xu^a, Da Chen^{a,*}^a State Key Laboratory of Precision Measurement Technology and Instruments, Tianjin University, Tianjin 300072, China^b College of Precision Instrument and Opto-Electronics Engineering, Tianjin University, Tianjin 300072, China

HIGHLIGHTS

- The DDRS enables the digital separation alternative to chemo-physical separation.
- The DDRS quantifies 12 gas components with dominant matrix interference.
- The DDRS method compared favorably with the GC method in realistic data sets.
- The limitation of detection of the DDRS ranged from 0.012% to 0.086%.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 21 April 2017

Received in revised form 8 June 2017

Accepted 9 June 2017

Keywords:

Natural gas

Raman spectroscopy

Gas sensing

Data driven

Higher-density discrete wavelet transform

High-throughput analysis

ABSTRACT

Mud logging system presents the most promising tool for exploring oil and gas resources, which relies on the multi-component analysis of gases to evaluate the hydrocarbon reservoirs. Higher detection efficiency, especially relating to the high-throughput analysis, can offer an important means to enhance the oil and gas recovery of hydrocarbon reservoirs. However, fewer technologies can measure both hydrocarbon and non-hydrocarbon gases in a timely manner. In the present work, a method based on data driven Raman spectroscopy (DDRS), which allows simultaneously accurate analysis of 12 hydrocarbon and non-hydrocarbon gases, has been proved. By developing a data driven strategy that incorporates higher-density discrete wavelet transform (HDWT) and template oriented frog algorithm (TOFA), the spectral resolution of Raman spectroscopy was significantly improved in both time and frequency domains to extract the target gas information from the Raman spectra, facilitating the detection of hydrocarbon reservoir. Theoretically, it is considered that the DDRS represents a promising tool for extracting the quantitative information from multi-component gases in the presence of dominantly uncontrolled variance. Our work has tested and refined the DDRS by performing a comprehensive analysis of three gas mixtures established to yield challenges representative of those encountered in a common mud logging analysis. The DDRS compared favorably with the gas chromatography (GC) method. The satisfactory results indicated that the DDRS has the capability to continuously detect 12 hydrocarbon and non-hydrocarbon gases in real time, which could be extended to online multi-component analysis of any other gas and fuel systems.

© 2017 Elsevier Ltd. All rights reserved.

1. Introduction

Natural gas is one of the most important fuel sources, and the exploration of natural gas relies on the continuous mud logging to detect hydrocarbon reservoir during drilling [1,2]. When the

* Corresponding authors.

E-mail addresses: qfli@tju.edu.cn (Q.-f. Li), dachen@tju.edu.cn (D. Chen).

drill bit crushes the rocks, both gaseous hydrocarbons and non-alkanes in the reservoir release from the rock pore space into the circulating drilling fluid [3]. The drilling fluid is then sent to a degasser to extract the gas mixture that includes methane, ethane, propane, iso-butane, normal butane, iso-pentane and normal pentane, as well as O_2 , N_2 , H_2 , CO_2 , CO for the evaluation of hydrocarbon reservoir [4,5]. The analysis of these 12 gases has been considered as a major challenge in the mud logging industry and many other industries [6]. For this reason, various gas detection techniques have been developed to enhance the oil and gas recovery.

Current analytical methods are mainly based on Gas Chromatography (GC) and its derived technologies [7,8]. However, the drilling speeds have reached the point that the current GC analyses of the evolved gases become a bottleneck for the mud logging industry [9]. In addition, GC is also difficult to detect both hydrocarbon and non-hydrocarbon gases simultaneously, i.e. 12 gases mentioned above. Moreover, GC instruments with special columns are rather expensive and bulky, requiring numerous maintenance work. Production-ready success demands incisive on-line methods to detect these 12 gases from drilling fluid.

A novel approach for simultaneous quantification of hydrocarbon and non-alkanes is Raman spectroscopy. This method offers a unique means to measure the gases nondestructively, with no sample preparation and little sensitivity to water vapor [10–13]. Since Raman spectroscopy can probe most of the molecular species simultaneously with a single light source, this technique is perfectly suitable for the multi-component analysis of gaseous fuels such as natural gas and biogas [14]. However, enhancement of the weak Raman signals is still a major challenge for Raman gas analysis.

An early feasibility verification of natural gas detection using Raman spectroscopy was reported by Diller and coauthors in 1980 [15]. After that, innovative efforts have been made by many researchers to achieve higher sensitivity and wider dynamic range of detection. Many novel methods, such as fiber-enhanced Raman spectroscopy (FERS) [16–18], cavity-enhanced Raman spectroscopy (CERS) [19–21], high power lasers [22] or high pressure cells [23], have been proposed to enhance the Raman signals. With these methods, a sub-parts per million detection limit for gas analysis can be obtained [23,24]. Meanwhile, the miniaturization of Raman gas analysis instrument represents another important advance of Raman techniques. Hippler and coauthors utilized CERS technique to achieve high sensitivity and excellent linearity of Raman spectroscopy with a 10 mW excitation laser [21], thus reducing the bulk of the Raman instrument efficiently. A portable Raman gas analysis system was also successfully developed for monitoring the gas components generated from a gas turbine power plant [25] and a biogas plant [26].

The advances of Raman spectroscopy have made the technique suitable for process analysis and onsite applications [27–30]. However, highly overlapped spectral bands and matrix interference combine to limit the applications of Raman spectroscopy. This creates a need for development of chemometrics methods tailored to encode certain quantitative signatures in the presence of overlapped Raman spectral interference.

Raman spectra are inherently multi-resolution in nature, consisting of spectral bands distributed in both frequency (peak width resolution) and time (wavelength position) domains [31]. Accordingly, the multi-resolution analysis provided by discrete wavelet transform (DWT) might be considered as a reasonable strategy to encode the analytical information in the presence of the complicated spectral interference [32,33]. With DWT, the multi-resolution analysis in frequency domain could eliminate the spectral background and noise accurately, while the multi-resolution analysis in time domain could isolate the spectral bands related

to analytes from matrix interference [34–36]. However, the down-sampling operations in DWT downgrade the spatial resolution in time domain, worsening the peak characteristics of the target analyte. To overcome this well-known disadvantage of DWT, oversampling strategy is proposed in wavelet transform to improve both time and frequency resolutions recently [37,38], thus extracting the quantitative information of the target analyte more efficiently.

A higher-density discrete wavelet transform (HDWT), based on a strategy of oversampling, has been proved to distinguish some overlapped bands with the improved spectral resolution [39,40]. HDWT differs from the conventional DWT in the way that it oversamples both time and frequency domains by a factor of two, resulting in a three times higher-density plane in dual domains. This arrangement leads to an expansive wavelet transform that processes intermediate scales with vanishing moments, thus sharpening the spectral features efficiently to promote the spectral resolution. With the combination of the feature selection strategies [41–44], an optimal subset of HDWT coefficients can be found with minimum overlap between the interested analytes and the other matrix substrates, facilitating the construction of a high quality calibration model.

The present work aims to explore the applicability of HDWT for encoding the interested bands in the presence of spectral interference, thus optimizing the prediction accuracy of quantitative calibration model for gas detection. In this work, a method based on data driven strategy has been proposed, which guides the quantitative analysis by selecting an optimal subset of the HDWT coefficients from the available spectra. We denote this method as data driven Raman spectroscopy (DDRS), which incorporates HDWT and template oriented frog algorithm (TOFA). Instead of randomly selecting the HDWT coefficients with event probability employed in the conventional Random Frog algorithm [45,46], the TOFA assigns coefficients with predefined weights to avoid choosing unimportant variables and shrink the search space as well. Consequently, the DDRS does not require any prior knowledge to extract the most important features correlated to the individual gas information.

Practically, the DDRS first finds an optimal combination of spectral features with the minimum overlapping, and then constructs the high-quality calibration models for gas analysis. In the experiments, we applied the DDRS for high-throughput analysis of 12 principal hydrocarbon and non-hydrocarbon components in standard gas mixtures, together with two real mud logging data sets. The results has shown a good performance of the DDRS to quantify a gas in the presence of dominantly uncontrolled variance, providing an isolated feature correlating to the interested analytes efficiently. Particularly, the DDRS model established with the standard gas mixtures predicted the real gas logging data well, which compared favorably with the GC method in practical mud logging sites.

2. Theory and algorithm

2.1. Higher-density discrete wavelet transform

Similar to DWT, HDWT decomposes a Raman spectrum into localized components labeled by a scale, and each component at different scale represents the features of different wavelength widths contained in the raw Raman spectrum. HDWT oversamples both time and frequency domain by a factor of two, and the wavelet frame associated with this expansive transform has two generators, $\psi_i(t)$, $i = 1, 2$ [40,47]. HDWT is implemented by the three channel filter banks, which is illustrated in Fig. 1. Here, H_0 , H_1 , and H_2 are low-pass, band-pass, and high-pass filters, respectively. The first two channels are down sampled by two, while the third

Download English Version:

<https://daneshyari.com/en/article/4768470>

Download Persian Version:

<https://daneshyari.com/article/4768470>

[Daneshyari.com](https://daneshyari.com)