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Gas geochemistry: From conventional to unconventional domains

A B S T R A C T

Gas geochemistry is developing into a powerful tool to understand geological processes and affirm source origins of geo-fluids. Major and trace gases, including abundances and isotopes, have shown considerable application in natural gas systems. For example, progress in unconventional gases such as shale gas, tight gas, and extreme conditions in the deep oceans represent more emerging areas and application of these novel gas-related techniques. Examples where gas geochemistry continues to place key constraints on the origin and migration characteristics of natural gas, the P-T characteristics of fluids in both subaerial and deep geothermal reservoirs, and the dynamics of the accumulation cycles, to name but a few. This volume will reflect this diversity in scope and application of gas geochemistry, focusing on deeper and broader applications in unconventional domains of novel gas geochemical techniques and applications.

1. Introduction

The continuing success of gas geochemistry is predicated upon innovative approaches involved in their detection, collection, extraction, preparation and measurement, as well as the development of new modeling techniques to further aid interpretation and understanding. This volume aims to document recent advances in gas geochemistry, illustrating their unique geochemical characteristics and application in contemporary research themes, and to explore their application to emerging areas such as exploitation of unconventional hydrocarbon deposits.

The 12th and 13th International Conference on Gas Geochemistry held in Petras, Greece in August, 2013 and in Chengdu, China in August, 2015 represent a unique gathering of gas geochemists from across the geosciences spectrum where the common theme is to understand, exploit and probe the usefulness and limitations of gases in a variety of applications. The Special Issue of MPG 'Gas Geochemistry: from conventional to unconventional domains' will reflect this diversity in scope and application of gas geochemistry, focusing on deeper and broader applications in unconventional domains of novel gas chemical proxies.

2. Themes

The 23 papers of this volume can be grouped into 4 general themes with considerable overlaps to some extent. Firstly, shale gas and shale are the most popular target for gas geochemists and this volume contains 6 papers aimed at understanding the origin, nature and extent of this important unconventional resource, their relationship to geologic setting, as well as associated micro-structure of shale and gas adsorption mechanisms.

Duan et al. (2017) address the chemical and structural characteristics of thermally simulated kerogen and its relationship with microporosity using solid-state ^{13}C nuclear magnetic resonance (NMR), Fourier transform infrared (FT-IR), Raman spectroscopy (RS), stable carbon isotopes, and Rock-Eval pyrolysis. They find that aromatic cluster size and aromaticity increases with the increasing maturation, whilst alkyl carbon is rearranged and attached into the increasing aromatic cluster, which confirms the molecular sieve effect in the kerogen samples. Wang et al. (2017a,b,c) characterize the shale pore structure using the organic-rich shale samples from Lower Silurian are in the Middle Yangtze region, central China. They show organic pores exist in organo-clay complex, and micropores are related to organic matters while mesopores and macropores are probably related to clay matters, especially illite. Zhang et al. (2017a,b) present the molecular compositions and carbon/hydrogen isotopes of a time series of produced shale gas samples from Lower Silurian Longmaxi Formation (Fm.) collected from six production wells from the Weiyuan and Changning shale gas pilots in Sichuan basin, China. They report interesting findings such as small fluctuations of $\delta^{13}C1$ and $CH_4/(C_2H_6 + C_3H_8)$ during production, showing an adequate gas supply, a 2‰ difference in $\delta^{13}C_1$ of shale gases from the Weiyuan horizontal and vertical wells suggests vertical heterogeneity, as well as the $\delta^{13}C$ difference between the Weiyuan and Changning shale gases which is caused by gas mixing from oil and kerogen cracking. Cao et al. (2017) present noble gas (He, Ne and Ar) abundances and isotopes of Longmaxi shale gas in Sichuan Basin to study shale heterogeneity, gas mixing process, gas supply and gas production changes. Xu et al. (2017) compare differential fluid migration behavior and tectonic movement in Lower Silurian and Lower Cambrian shale gas systems in Sichuan, China using isotopic data. They reveal that isotopically reversed gas in Longmaxi shale implies a sealed and self-contained petroleum system while low pressure

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Cambrian Qiongzhusi shale gas suggests a tectonic destructed system. Luo et al. (2017) have studied the geological and geochemical characteristics of marine-continental transitional shale from the Upper Permian Longtan formation, Northwestern Guizhou, China, which have shown different pore structures and accumulation patterns from marine shale.

The second and strongly related theme is the topic of high-over mature marine gas and deep gas in China, which have successfully been explored in recent years. Zhang et al. (2017a,b) report the unique chemical and isotopic characteristics and origins of natural gases in the Paleozoic marine formations in the Sichuan Basin, SW China focusing on isotope fractionation of deep and high mature carbonate reservoir gases. They propose two quantitative mixing models, regarding the disappearance of the ^{13}C reversal caused by ethane cracking, as well as hydrogen isotopic exchange leading to D isotope rollover for methane. However, Qin et al. (2017a) propose another model to explain the isotopic characteristics for the same area. They suggest the less negative $\delta^{13}C_1$ values may be caused by the release of water-dissolved gas, and the geological setting of the Anyue gas field is favorable for the formation, preservation and release of water-dissolved gas. The other two papers shift from Sichuan Basin to other two basins in China: Tarim Basin and Ordos Basin. Liu et al. (2017) study the geochemistry characteristics and genetic types of natural gas in central Tarim Basin, and they show that oil-type gas experiences different formation processes of kerogen-cracking, oil-cracking, oil- and gas-cracking, secondary gas-cracking and mixing of the kerogen-cracking and oil-cracking gases, showing a trend from gas-cracking gas, to oil- and gas-cracking gas and oil-cracking gas along the direction from west Manjar Sag to the central Tarim Basin. Li et al. (2017a,b) examine the characteristics and genetic types of the Lower Paleozoic natural gas in Ordos Basin. Their results show that the oil-associated gas is closely related to Lower Ordovician source rocks, and CO_2 and N_2 are mainly come from Ordovician whilst coal-derived gas is derived from C-P coal measures. Another paper by Mi et al. (2017) report a laboratory experimental result of the upper thermal maturity limit of primary gas generated from marine organic matters. The propose a model that the primary gas generation from marine OM mainly occurs below 2.0 %Ro and the upper thermal maturity limit might extend to about 3.5 %Ro but with less contribution (<10%) of gases.

The third category mainly focuses on tight gas, coal gas and shale oil, which is also a very important domain in unconventional gas exploration. Wu et al. (2017) report a study on geochemical characteristics of tight gas and its source interpretation in the Daniudi gas field, the Ordos Basin, China. The molecular and methane carbon isotopic fractionations of natural gas suggest that the Daniudi tight gas is coal-derived gas, and the lower Permian (P_{1s}-P_{1x}) and Upper Carboniferous (C_{3t}) gas were derived from their own source rocks, respectively. Qin et al. (2017b) propose that water-soluble gas might play a significant role in formation of the tight coal-derived-gas reservoirs with medium-low abundance in Xujiahe Formation, central Sichuan Basin, China. Wang et al. (2017a,b,c) report the results of molecular and isotopic compositions of noble gases and hydrocarbon gases in the Kela 2 gas field, Tarim Basin, China and infer their origins comprehensively. Liao et al. (2017) conduct a kinetic experiment of marine and lacustrine shale grains using Rock-Eval pyrolysis and find shale grains exhibit a broader distribution of activation energy (Ea) and higher dominant Ea than powder and kerogen, and enjoy a higher expulsion threshold and higher retention ability. They also point out that Yanchang Lacustrine shale in the Ordos basin shows a stronger retention than Pingliang marine shale, suggesting higher shale oil prospects.

The fourth category is broad in scope, including abiogenic gas and chemical reactions in its formation process as well as gas hydrate and gas seepage subaerially in different areas. D'Alessandro et al. (2017a) present the analytic results of gases collected from dry seepages and gases both bubbling and dissolved in springs of the ophiolitic body of Kizildag (Turkey). They discover large compositional variation (H_2- CH_4 or N_2-dominated) and large isotopic fractionation of methane at one site due to biological oxidation, which implies hydrogen derives from low temperature serpentinization processes, and methane mainly derives from abiotic processes. In another case study, Argolida ophiolite (Ermioni, Greece), D'Alessandro et al. (2017b) report a newly discovered hyperalkaline spring with typical Ca-OH composition where methane is mainly derived from abiotic serpentinization processes. Ji et al. (2017) identify and group the mixed sporopollen assemblage from sediments of the Dushanzi mud volcano in southern Junggar Basin, suggesting that the Dushanzi mud volcano eruption is related to damage of gas reservoirs since the Neogene. Baciu et al. (2017) assess the hydrocarbon seeps in Romania and measure the flux released into the atmosphere. They show that Transylvanian gases are mainly microbial while those in the East Carpathians and Moldavian Platform are mainly thermogenic. The total methane output from 94 seeps is estimated at least 3000 t y^{-1}. Cheng et al. (2017) report on hydrocarbon sources for oil gas associated with gas hydrate in the Qilian Mountain permafrost, Qinghai, Northwest China. They identify two types of oil gas indicators and conclude the gas hydrate has a similar parent biomass as the Middle Jurassic source rocks and originated from deeper source rocks than the Middle Jurassic. Ma et al. (2017) report an experimental result of minerals and iron on natural gas generation during pyrolysis of type-III kerogen and they find mixed geological catalysts improve much more gaseous hydrocarbon yields.

This volume also includes two technical papers: the first by Wang et al. (2017a,b,c) who propose an alternative method to estimate volumetric vapour: liquid ratio of gaseous or light-petroleum inclusions without fluorescence, using regularly-shaped inclusions through geometric methods, and the other is by Li et al. (2017a,b) who develop a novel method for D/H ratio measurements for volatile hydrocarbons of crude oils using solid phase micro-extraction (SPME) coupled to gas chromatography isotope ratio mass spectrometry (GC-IRMS). These two techniques have been proved both effective and applicable.

3. Concluding remarks

The ICGG series of meetings provided a lively forum for innovative ideas and broad discussions focused exclusively on gas geochemistry. The ICGG 13 chose Chengdu city, Sichuan mainly because of its long history of gas exploitation and diverse natural gas types, its special geographical proximity to the boundary location of the Tibetan plate, abundant geological phenomena related to natural gases, massive earthquake relics as well as its unique culture. This meeting attracted around 120 delegates. The four-days meeting included 53 oral presentations and 57 posters covering 7 themes related to geochemistry. This meeting set up a special session in memorial of Professor Tsan-yao F. Yang for his contribution to the ICGG community. A pre-conference workshop on noble gas geochemistry was organized, and two distinguished scientists, Prof. Yuji Sano form University of Tokyo and Prof. David R. Hilton from Scripps Institution of Oceanography, University of California, San Diego gave two lectures to about 60 young geochemists from the world. A four-day post-conference field trip was organized allowing delegates to experience the unique geological characteristics and diverse nature of gas phenomena, and likewise the special natural and cultural attractions of Sichuan and Tibet. Delegates observed the active tectonics of the Tibetan Plateau and the adjacent region in addition to the spectacular earthquake surface

rupture zones related to the 2008 Ms 8.0 Wenchuan earthquake. Seismic relics, induced by large earthquakes related to geological disasters in the Longmen Shan Mountains were also identifiable. Delegates also visited the massive travertine landscape in Songpan, as well as an outcrop of the Cambrian black shale which is the present source of shale gas exploitation in the Sichuan Basin. Proceedings of the meetings are traditionally published as Special Issues of peer-reviewed journals, as in the present case. ICGG continues to attract widespread participation – in the form of oral and poster presentations - from a diverse international audience. We look forward to the 14th ICGG scheduled to be held in Poland in September, 2017.

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