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New indexes and charts for genesis identification of multiple natural gases

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Abstract: Identification of natural gas genesis and source for high-matured multiple natural gases is a great challenge in the exploration of deep–ultra deep and unconventional natural gases. In this paper, the genesis identification method system of multiple natural gases is enriched through new experimental techniques and comprehensive analysis of geological data. New indexes and charts of genesis identification for multiple natural gases were determined to distinguish the sapropelic kerogen degraded gas and oil cracking gas, accumulated and scattered liquid hydrocarbon cracking gas in different evolution stages, nitrogen, carbon dioxide of organic and inorganic origins, inert gases of crustal and mantled origins, coal-formed gas and oil-typed gas by helium, nitrogen, carbon dioxide and mercury content in natural gas. These indexes and charts have been successfully applied in the Sichuan, Tarim and Songliao basins to identify the natural gas genesis and source for complicated gas reservoirs. The research results have provided effective supports for the natural gas exploration in the Sinian–Cambrian ancient carbonate formations in the Sichuan Basin, deep formations in the Kuqa depression of the Tarim Basin, and deep volcanic formations in the Songliao Basin.

Key words: multiple natural gases; genesis identification; cracking gas; nitrogen; carbon dioxide; inert gas; mercury content

Introduction

Natural gas is composed of methane, ethane, propane, butane, C₅₋₈ light hydrocarbons, nitrogen, carbon dioxide, and inert gases etc. As natural gas is simple in composition, with little usable information and few indicators, it is difficult to find out the origin of natural gas. Galimov, Stahl, Welhan, Dai Jinxing, Xu Yongchang, Liu Wenhui and other researchers carried out studies on identification of organic and inorganic origins of natural gas early^[1-6], pushing the progress of natural gas origin identification. Especially since "the 6th five-year plan" in China, the team led by academician Dai Jinxing has set multiple identification indexes and charts of four categories, natural gas component, carbon isotope, light hydrocarbon, and biomarker, for identifying coal-derived gas and oil-typed gas^[4,7-11], which have provided crucial support for identifying natural gas origin and guiding coal-derived gas exploration in China.

In recent years, with natural gas exploration extending toward deep to superdeep formations and unconventional domain, and the discovery of multiple kinds of complex gas reservoirs, such as oil cracking gas (including accumulated and scattered liquid hydrocarbon cracking gas), high nitrogen and high carbon dioxide gas reservoirs, the existing natural gas origin identification methods can't distinguish sapropelic kerogen degraded gas from oil cracking gas, accumulated liquid hydrocarbon cracking gas from scattered liquid hydrocarbon cracking gas at high thermal evolution stage, and gas of organic origin from gas of inorganic origin, etc, which are crucial for the identification of origin and source of high evolution degree and complex gas reservoirs. Since the information usable in natural gas is limited, making full use of hydrocarbons, helium, nitrogen, carbon dioxide, and mercury composition data seems especially crucial. With the help of geochemical test methods and newly developed techniques, new indexes and charts have been established to identify various natural gases, such as the sapropelic kerogen degraded gas and oil cracking gas, accumulated and scattered liquid hydrocarbon cracking gas with different thermal evolution stages, N₂ and CO₂ of organic and inorganic origins, inert gas of crustal and mantled origins, coal-formed gas and oil-typed gas by He, N₂, CO₂ and mercury content in natural gas; and the

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origins and sources of deep high evolution degree and complex gases in some major gas-bearing basins have been explored in this study, in the hope to guide gas exploration there.

1. Identification of oil cracking gas

1.1. Distinguishing sapropelic kerogen degraded gas and oil cracking gas

The origin of the old carbonate gas fields with deep burial depth and high thermal evolution is complicated. It could come from the primary degradation of kerogen, or from secondary cracking of oil. Based on thermal evolution simulated experiments of types II and III kerogen, Prinzofer et al^[12], Behar et al^[13] set up the identification chart of kerogen degraded gas and oil cracking gas, but these experiments were done at lower thermal evolution degree, and didn't consider the effect of temperature rise rate on parameters fully, so the identification chart can't distinguish kerogen degraded gas in high evolution stage from oil cracking gas effectively. Therefore, low maturity sapropelic shale samples (TOC=2.79%, R_o =0.52%) from the Xiamaling Formation of Neoproterozoic Qingbaikou System were taken, gas generation simulated experiments on the original kerogen in this shale, oil (liquid hydrocarbon generated by heating the original kerogen to oil generation peak in conventional autoclave thermal simulation unit), and residual kerogen (residual sample after the liquid hydrocarbon was removed) were conducted in high temperature high pressure gold tube system and autoclave thermal evolution simulated equipment, and the products of the experiments were analyzed. Finally the $ln(C_1/C_2)-ln(C_2/C_3)$ chart for identifying sapropelic kerogen degraded gas and oil cracking gas with different thermal evolution stages has been set up (Fig. 1).



Fig. 1. Identification chart for sapropelic kerogen degraded gas and oil cracking gas of different evolution stages.

In the chart, the red curve denotes the variations of oil cracking gas $\ln(C_1/C_2)$ and $\ln(C_2/C_3)$ with maturity, and the blue curve denotes the variations of kerogen degraded gas $\ln(C_1/C_2)$ and $\ln(C_2/C_3)$ with maturity. It can be seen the two kinds of gases have apparently different evolution features: the $\ln(C_2/C_3)$ of oil cracking gas increases rapidly in the early stage and is basically stable in late stage; while the $\ln(C_2/C_3)$ of kerogen degraded gas shows a variation trend of stable-fast increase-stable-fast increase. The above difference is probably related to the structure of oil and kerogen, the activation energy needed for cracking and degradation, and the productivity of hydrocarbon gas.

Sinian-Cambrian gas in Gaoshiti-Moxi area of the Sichuan Basin (simplified as Gao-Mo area) has a $\ln(C_1/C_2)$ range of 6.35–7.85 and $\ln(C_2/C_3)$ range of 3.11–4.69, basically falling in the scope of oil cracking gas with R_o of over 2.5% in the chart, indicating the Sinian-Cambrian gas is primarily oil cracking gas (Fig. 1 and Table 1). This understanding agrees with the facts that there is rich carbonaceous asphalt left after oil cracking in the reservoir and the light hydrocarbons in this gas shows the features of oil cracking gas^[14-16]. In contrast, those values of gas samples from the Xujiahe Formation in central Sichuan Basin fall in the scope of kerogen degraded gas with R_o of 1.0%–2.5%, indicating the Xujiahe Formation gas is kerogen degraded gas. In addition, gas samples from the Carboniferous in eastern Sichuan Basin, Well Zhongshen1 Cambrian in the Tarim Basin, the Ordovician in Hetianhe Gas field and Lunnan Gas field all fall in the scope of oil cracking gas with R_0 of 1.5%–2.5%, indicating they are all oil cracking gas.

1.2. Identification of accumulated and scattered liquid hydrocarbon cracking gas

Oil cracking gas is a major kind of gas in petroliferous basins, including accumulated liquid hydrocarbon cracking gas originated from ancient oil reservoir and scattered liquid hydrocarbon cracking gas retained in source rock and migration pathway^[17]. Methyl cyclohexane/n-heptane index for identifying accumulated and scattered liquid hydrocarbon cracking gas was established ^[17], but based on oil cracking experiment in single medium (montmorillonite), the index needs to be improved further. In this study, by using gold tube confining system simulated equipment, cracking simulated experiments with Ordovician marine oil samples from Well Lunnan32 at different oil/medium ratios (80% oil + 20% medium, 50% oil + 50% medium, 30% oil + 70% medium, 15% oil + 85% medium, 5% oil + 95% medium, 2% oil + 98% medium, and 1% oil + 99% medium) and temperatures (370 °C, 385 °C, 400 °C, 415 °C, 430 °C, 445 °C, 460 °C, 475 °C, 490 °C) with different media (calcium carbonate, montmorillonite) have been conducted, and the characteristics and differences of C_6 — C_7 in oil cracking gases of different cracking degrees and state have been examined carefully (Fig. 2). The experiment results show the $\sum C_6 - C_7$ cycloalkane/(nC₆+nC₇) and methyl cyclohexane/

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