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Apatite fission-track and Re-Os geochronology of the Xuefeng uplift, China: Temporal implications for dry gas associated hydrocarbon systems

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ABSTRACT

Hydrocarbon evolution is extremely challenging to determine, both temporally and spatially, in complex tectonic settings. Here we investigate the western margin of the Xuefeng uplift (southern China), which records multiple and protracted tectonic and hydrocarbon-generation events. This timing of initial oil generation is recorded by low-maturity bitumen (type A), which yields an Re-Os bitumen date of ca. 430 Ma, consistent with basin models and a ca. 405 Ma bitumen Rb-Sr date. In contrast, apatite fission-track (AFT) data yield considerably younger dates that reflect the timing and tectonic evolution of the Yanshan orogeny from the northwest (ca. 150 Ma) to the southeast (ca. 70 Ma). The youngest AFT date coincides with the western margin of Xuefeng uplift, where high-maturity bitumen (type B) occurs that yields a ca. 70 Ma Re-Os date. The Re-Os and AFT dates imply that both the last stage of the Yanshan orogeny and, by inference, the cessation of dry gas generation, occurred ca. 70 Ma. The Re-Os data of this study imply that the Re-Os chronometer can aid in constraining the timing of oil generation and secondary and/or more mature hydrocarbon processes (e.g., thermal cracking and/or gas generation) in hydrocarbon systems worldwide.

INTRODUCTION

Being able to accurately constrain the timing of hydrocarbon generation is vital for understanding the evolution of a petroleum system and for hydrocarbon exploration. Although the timing of oil generation can be estimated by numerous methods, establishing the timing of gas formation during hydrocarbon evolution is currently less definitive (Schaefer, 2005). To date, basin modeling and hydrocarbon fluid inclusion analysis have been widely used to evaluate the timing of gas generation from oil cracking (Roberts et al., 2004). However, imperfect kinetic models and poorly constrained parameters, such as the paleo–geothermal gradient, pressure conditions, and the physical properties of the strata, hamper the accuracy of any derived age model (Braun and Burnham, 1992). Although studies are limited (Qiu et al., 2011), quartz fluid inclusion Ar-Ar dating has shown potential to yield the timing of gas emplacement; however, this method is hampered by requiring abundant gas-liquid inclusions and the challenging analytical protocol (Liu et al., 2011; Qiu et al., 2011). Apatite fission-track (AFT) dating has also proven useful, because it can directly track the thermal history of a sedimentary basin (Donelick et al., 2005). Hitherto, the rhenium and osmium (Re-Os) isotope system has provided temporal constraints for oil generation and migration (Lillis and Selby, 2013; Selby and Creaser, 2005), but without any reference to the timing of gas formation. In this study we apply both AFT and Re-Os geochronology to the Majiang and Wanshan reservoirs of the Xuefeng uplift, southern China, to quantitatively constrain the timing of tight gas generation, which is currently highly debated (Schenk et al., 1997).

GEOLOGICAL SETTING

The Majiang and Wanshan hydrocarbon reservoirs are the two largest in southern China, containing a reserve exceeding 10 bbl (billion barrels) of oil and 200 bcm (billion cubic meters) of gas (Deng et al., 2014; Wu, 1989) (Fig. 1). The gas reserve is close to that of the second-largest gas field in China, Puguang (Ma et al., 2007), and the Ledovoe gas field in Russia (Pavlenko and Glukhareva, 2010), which each contain ~300 bcm of gas. The Majiang and Wanshan reservoirs are located in the foreland basin belt along the western margin of the Xuefeng uplift of the mid-Yangtze block (Deng et al., 2014) (Fig. 1). The foreland basin is characterized by multiple tectonic events (the Paleozoic Caledonian, Triassic Indosinian, and the Cretaceous–Holocene Yanshan-Himalaya orogenies; Mei et al., 2012); this has resulted in numerous tectonic models and timing constraints for hydrocarbon generation (Bai et al., 2013; Liu et al., 2011; Tang and Cui, 2011). Oil and bitumen generation from Paleozoic strata is proposed to have occurred during tectonic subsidence associated with the late Silurian Caledonian and Triassic Indosinian orogenies (Bai et al., 2013; Liu, 2011). However, the timing of gas generation is less certain, with estimates tied to both the Triassic Indosinian and Cretaceous Yanshan orogenies (Liu, 2011; Xiang et al., 2008). For example, a ca. 228 Ma date derived from Ar-Ar analysis of hydrocarbon-bearing fluid inclusions in the Majiang reservoir has been used to suggest that hydrocarbon generation of both oil and wet gas was contemporaneous with the Indosinian orogeny (Liu, 2011). In contrast, basin modeling coupled with gas fluid inclusion analysis suggests that dry gas formation occurred during the Cretaceous (Xiang et al., 2008); this coincides with the Yanshan orogeny, constrained by AFT dates (ca. 92 Ma) from ~100 km to the north of the study area, and quartz electron spin resonance (ESR) dates (75–62 Ma) along the Jiangnan fault zone in the Xuefeng uplift, ~500 km to the east of the study area (Mei et al., 2010; Zhu et al., 2011) (Fig. 1).

Bitumen is commonly hosted within the pore spaces of Cambrian to Ordovician limestone units, but is also present along fractures and cleavage planes of Silurian sandstone units (Zhou, 2006). Organic geochemistry defines two types of bitumen in the Majiang and Wanshan reservoirs. In the Kaili area only, which represents a structural high, of the Majiang reservoir (Fig. 1), bitumen (type A) has low maturity (reflectance, Ro < 1.0), low temperature, T_{max} (~450 °C), yellow fluorescence, and a high H/C ratio (>0.8) (Han et al., 1982; Zhou, 2006). In contrast, bitumen (type B) throughout the rest of the Majiang and Wanshan reservoirs (Fig. 1) has high maturity (Ro > 2.0), high T_{max} (~550 °C), a low H/C ratio (<0.6), no fluorescence, and contains high adamantane (3, 4 diamondoids [3, 4-DMA]) concentrations (~50–300 ppm); this classifies the bitumen as pyrobitumen (Shi et al., 2015; Wang et al., 2013). Though several processes can form pyrobitumen (Lewan, 1997),

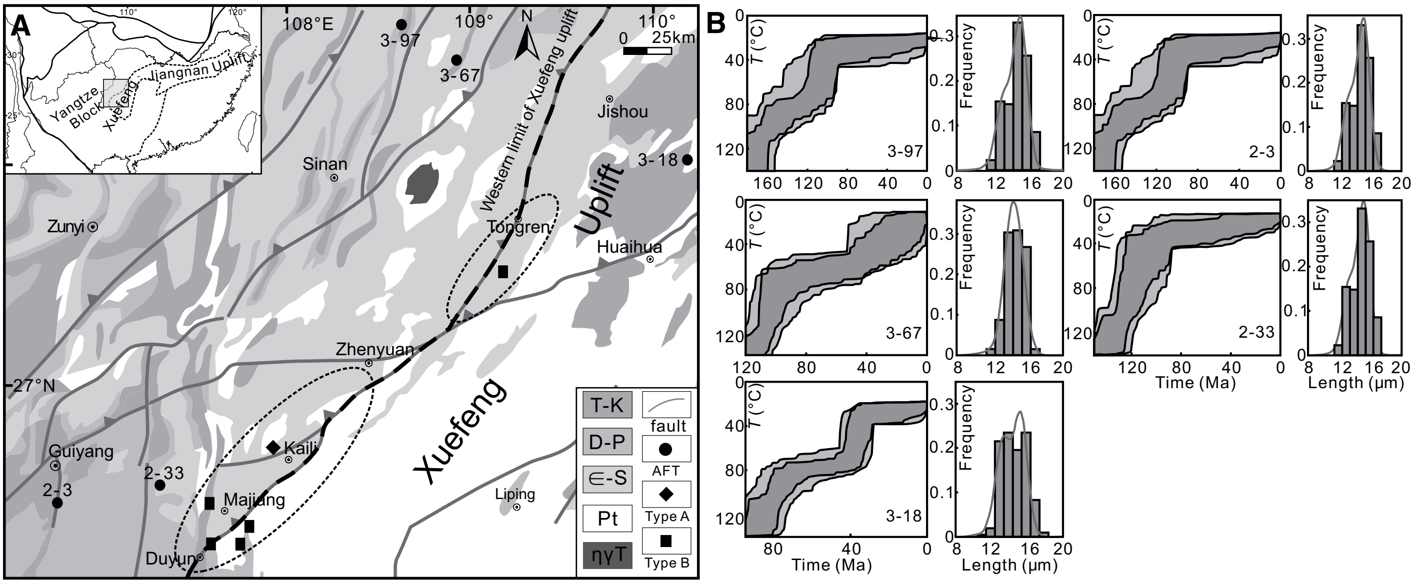

Figure 1. A: Simplified geological map of western margin of Xuefeng uplift (southern China) showing the locations of the type A and type B bitumen in the Majiang and Wanshan reservoirs, and sandstone samples for Re-Os and apatite fission track (AFT) analysis. T-K—Triassic to Cretaceous; D-P—Devonian to Permian; ∈-S—Cambrian to Silurian; Pt—Precambrian; ηγT—volcanic rock. B: Thermal history inversions from AFT data for sandstone samples using HeFTy software (Table DR1; see footnote 1). See text for discussion.

dry gas and pyrobitumen can form contemporaneously by thermal cracking of low-maturity bitumen (e.g., type A of this study) and crude oil (Huc et al., 2000). To evaluate the timing of both bitumen types of the Majiang and Wanshan reservoirs, and by inference the timing of oil and gas generation, we conduct AFT analyses on the Paleozoic and Jurassic sandstones, and Re-Os analyses on both type A and B bitumen.

SAMPLES AND METHODOLOGY

Samples for AFT analysis were collected from the Ordovician Dawan Formation (sample 2-33; Majiang reservoir), Jurassic Ziliujing Formation (sample 2-3; Majiang reservoir), Silurian Shiniulan Formation (sample 3-67; Wanshan reservoir), Triassic Guanling Formation (sample 3-97; Wanshan reservoir), and the Triassic Xiaojiangkou Formation (sample 3-18; Xuefeng uplift) (Fig. 1A; Table DR1 in the GSA Data Repository[1]). Three samples (samples 3-97, 3-67, and 3-18) were broadly collected along a northwest-southeast profile in the north of the study area, parallel to the propagation direction of the Yanshan orogeny. Samples 2-33 and 2-3 were collected ~200 km to the southwest of samples 3-97, 3-67, and 3-18 (Fig. 1A).

Type A and B bitumen were sampled from outcrops across the Majiang and Wanshan reservoirs (Fig. 1A; Table DR2). Five type A bitumen samples, only known in outcrops in the Majiang reservoir, in an ~20-m-long exposure in the Ordovician Honghuayuan Limestone Formation ~20 km west from the city of Kaili, were collected (Fig. 1A). In contrast to type A bitumen, type B bitumen is widely found, and predominantly occurs in a northeast-southwest–trending belt in early Paleozoic limestone in the Majiang and Wanshan reservoirs west of the Xuefeng uplift (Fig. 1A). Nine samples were collected from the Ordovician Honghuayuan Formation in the Majiang reservoir. Samples MJ-S1-B, MJ-S2-B, and MJ-S4-B were collected with an ~10 m interval spacing from an ~30 m section ~2 km south of the city of Majiang. Sample XR-S1-B is from ~3 km west of the town of Xingren. Samples HBZ-S1-B, HBZ-S4-B, HBZ-S6-B, and HBZ-S7-B are from a 20 m section, collected ~4 m apart, ~1 km northeast of the town of Huoba. Sample MJ1-2 was collected from ~1 km northeast of the town of Pojiao. In addition, three samples were collected from the Cambrian Aoxi Formation in the Wanshan reservoir. Samples WS-S3-B, WS-S4-B, and WS-S4-B6 were collected with an ~5 m interval spacing from an ~30 m section ~3 km from the city of Wanshan (Fig. 1A). (For the detailed analytical protocol and data for both the AFT and Re-Os analysis, see the Data Repository.)

RESULTS

The AFT data for the samples collected along a profile parallel to the propagation direction of the Yanshan orogeny yield dates, from northwest to southeast, of 155 ± 13 Ma (sample 3-97), 97 ± 7 Ma (sample 3-67), and 71 ± 6 Ma (sample 3-18) (Fig. 1A; Table DR1). The two samples in the southwest of the study area yield AFT dates of 150 ± 7 Ma (sample 2-3) and 123 ± 7 Ma (sample 2-33) (Fig. 1A; Table DR1). The mean track lengths of all samples are 12.66–13.71 μm (1 standard deviation = 1.43–2.21 μm; Table DR1). Thermal history modeling of the AFT data shows continuous cooling from ca. 160 to 70 Ma from northwest to southeast (Fig. 1).

Type A bitumen samples have 1.5–4.3 ppb Re and 76.4–206.4 ppt (parts per trillion) Os, with $^{187}Re/^{188}Os$ and $^{187}Os/^{188}Os$ compositions of 113–121 and 1.60–1.66, respectively (Table DR2). The type A bitumen Re-Os data yield a model 1 (which assumes that the assigned uncertainties are the only reason for any scatter in the fit of the data) date of 429 ± 140 Ma, with an initial $^{187}Os/^{188}Os$ (Os_i) value of 0.79 ± 0.27 (mean square of weighted deviates, MSWD = 0.41) (Fig. 2A). Type B bitumen has typically higher Re (2.5–15.2 ppb) and Os (40.0–498.1 ppt) abundances, and have a greater variability in their $^{187}Re/^{188}Os$ and $^{187}Os/^{188}Os$ compositions (87–497 and 1.52–1.97, respectively; Table DR2). The type B bitumen Re-Os data yield a model 3 (which assumes that the scatter in the degree of fit of the data is a combination of the assigned uncertainties, plus a normally distributed variation in the $^{187}Os/^{188}Os$ values) date of 69 ± 24 Ma (Os_i = 1.45 ± 0.09, MSWD = 9.6) (Fig. 2B).

DISCUSSION AND IMPLICATIONS

The Xuefeng uplift and adjacent districts to the west are part of a piggyback thrust system that has a northwest transport direction (and therefore youngs toward the southeast) and developed from the late Mesozoic, as a result of the Pacific-Yangtze plate collision (Yanshan orogeny; Yan et al., 2003) (Fig. 1A). Restoration of the section across the Majiang reservoir shows

[1]GSA Data Repository item 2016163, methods and data, is available online at www.geosociety.org/pubs/ft2016.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

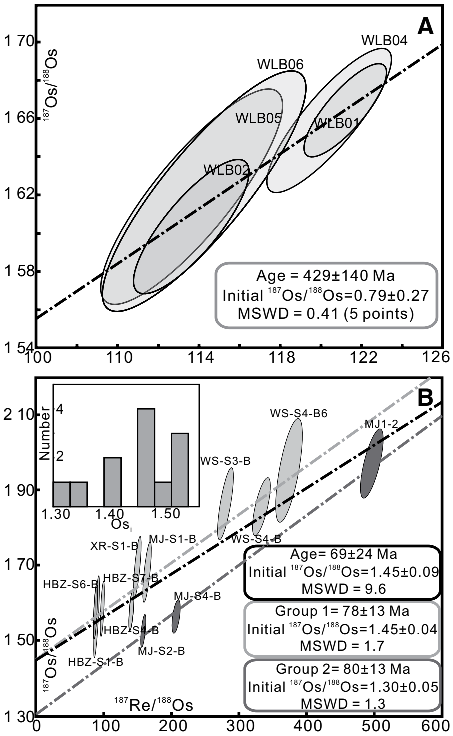

Figure 2. A: ^{187}Re-^{187}Os isochron for five type A bitumen samples from Kaili area, Majiang reservoir (southern China). MSWD—mean square of weighted deviates. B: ^{187}Re-^{187}Os isochron diagram for 12 type B bitumen samples from Majiang and Wanshan reservoirs. All data include 2σ uncertainties and the error correlation function, ρ (Table DR2; see footnote 1). Os_i—initial $^{187}Os/^{188}Os$. See text for discussion.

that ~20% (~12 km) shortening occurred during the Cretaceous (Zhang, 2010). The AFT dates (ca. 150–70 Ma) obtained in this study coincide with the estimated timing of the Yanshan orogeny and support the southeast younging direction of the thrust system (Mei et al., 2010; Zhu et al., 2011). For example, samples 2-3 and 3-97 from 150 km to the northwest of the Xuefeng uplift belt have similar AFT dates of ca. 150 Ma. Significantly younger AFT dates (ca. 120 and ca. 100 Ma) were obtained for samples 2-33 and 3-67 from ~90 km northwest of the Xuefeng uplift. An AFT date of 92 Ma was also obtained from ~50 km northwest of Xuefeng uplift (Mei et al., 2010). The ca. 70 Ma AFT date (sample 3-18) recorded within the Xuefeng uplift (Fig. 1A; Table DR1) is similar to the ESR dates (75–62 Ma) for the northern part of the Xuefeng uplift (Zhu et al., 2011). The AFT dates indicate that the Yanshan orogeny was a prolonged (ca. 90–70 Ma) tectonic event, with the last phase occurring ca. 70 Ma.

A thermal history derived from the AFT lengths using the HeFTy software (version 1.8.1) and employing a fanning curvilinear annealing model (Ketcham et al., 2009), with the parameters of (1) the annealing temperature (60–120 °C) of the apatite fission tracks, and (2) a present-day surface temperature of 20 °C, yields the same outcome as that determined by the AFT dates; e.g., uplift events ca. 150, ca. 100, and ca. 70 Ma from the northwest to the southeast across the western margin of the Xuefeng uplift (Fig. 1B; Table DR1). The AFT dates and thermal history model also yield, by inference, a time frame for the cessation of hydrocarbon generation. Given that the thermal cracking of oil and bitumen requires temperature of ≥150 °C (Huc et al., 2000), the spatial association of type B bitumen and gas within and/or near the Xuefeng uplift suggests that hydrocarbon generation had ceased by ca. 70 Ma, during the final stage of the Yanshan orogeny.

A late Silurian–Early Devonian Re-Os date is recorded for type A bitumen (429 ± 140 Ma) (Fig. 2A). Although having a large uncertainty, likely because of the low Re and Os abundance and limited spread in $^{187}Re/^{188}Os$ and $^{187}Os/^{188}Os$ values, the nominal date calculated by the Re-Os data for type A bitumen broadly coincides with the timing for the initial oil generation along the western margin of the Xuefeng uplift in the Majiang reservoir, based on burial models of the basin, oil-bearing fluid inclusions with homogenization temperatures of ~100 °C (Bai et al., 2013), and Rb-Sr bitumen geochronology that yields an age of 405 ± 20 Ma (Tang and Cui, 2011).

In agreement with the ca. 70 Ma AFT date is the Re-Os date (69 ± 24 Ma, MSWD = 9.6) for type B bitumen (Fig. 2B). Type B bitumen has geochemical characteristics different from those of type A bitumen; e.g., type B bitumen has no fluorescence and contains adamantane, indicating that the bitumen is pyrobitumen. Pyrobitumen could form by the thermal cracking of bitumen and/or oil at temperatures >150 °C (Huc et al., 2000). Along the western margin of the Xuefeng uplift, continuous subsidence since the Late Triassic led to the burial of Paleozoic strata to a depth of ~5000 m, with temperatures >150 °C being attained during the Late Jurassic to Early Cretaceous (Bai et al., 2013; Han et al., 1982). At this temperature, coincident with the formation of pyrobitumen, gas dominated by methane, which is found in the Majiang and Wanshan reservoir, also formed (Zhou, 2006). Although spatially close to the type B bitumen (~40 km), type A bitumen in the Kaili area did not undergo thermal cracking because it occurs structurally higher on the footwall of the Shanban fault (Fig. 1A), and as a result, only underwent temperatures associated with oil generation during the late stages of the Caledonian orogeny (latest Silurian) (Zhang, 2010).

Our AFT dates indicate that the Yanshan orogeny caused the uplift of the Majiang and Wanshan reservoirs to a level where the ambient temperature was between 120 and 60 °C by ca. 70 Ma. The AFT dates, coupled with basin burial models, fluid inclusion homogenization temperatures, and hydrocarbon composition numerical modeling (evolution of C_{14+} to C_1 compounds with time), suggest that thermal cracking of hydrocarbons (e.g., type A bitumen and oil) may have occurred over 75 m.y. (Late Jurassic to Late Cretaceous) (Huc et al., 2000; Xiang et al., 2008). Although the Re-Os date for type B bitumen agrees with the AFT dates, the statistical fit of the Re-Os data yields a large MSWD of 9.6 and a large date uncertainty (34%), which suggests that the Re-Os data have not fully met the criteria to obtain a precise isochron. For example, the requirements to yield a precise isochron are that the sample set represents contemporaneous formation, has identical initial isotope compositions (IOs), and that the Re-Os systematics have not been disturbed. Nevertheless, the agreement of the AFT and Re-Os chronometers suggests that the process of thermal cracking has reset the Re-Os systematics in the thermally cracked type A bitumen. As such, the Re-Os type B bitumen date is recording the end of pyrobitumen formation and, by inference, the cessation of dry gas generation. The agreement of the AFT and Re-Os type B bitumen dates may also indicate that the Re-Os systematics in hydrocarbons have a closure temperature range similar to that of AFT (e.g., 120–60 °C; Kohn and Green, 2002). This range also coincides with the temperature condition (~100–140 °C) of thermochemical sulfate reduction (Machel, 2001), a process that has also been proposed to disturb or even reset the Re-Os isotope systematics in oil (Lillis and Selby, 2013). Assuming no significant disturbance to the Re-Os systematics of the type B bitumen since the uplift related to the Yanshan orogeny, the scatter in the data and the large date uncertainty may be controlled by the IOs of the type B bitumen sample set.

Calculating IOs values for the samples at 70 Ma results in a range from 1.32 to 1.51 (Table DR2) that is broadly defined by two groups (group 1 = 1.32–1.39, n = 3; group 2 = 1.42–1.53, n = 9; Fig. 2B). Treated as two groups, the Re-Os data yield identical, but more precise (±16%) model 1 Re-Os dates (group 1 = 80 ± 13 Ma, IOs = 1.30 ± 0.05, MSWD = 1.3; group 2 = 78 ± 13 Ma, IOs = 1.45 ± 0.04, MSWD = 1.7) (Fig. 2B). Although more precise, the Re-Os dates are still in agreement with the ca. 70 Ma AFT date and AFT thermal modeling in the Xuefeng uplift. The coupled AFT and pyrobitumen Re-Os dates both record the last stage of tectonic uplift of the Xuefeng uplift and yield the best estimate for the cessation of dry gas formation. The long-lived Yanshan orogeny controlled the final evolution of the hydrocarbon system and led to the near surface exposure of pyrobitumen. Uplift, coupled with erosion, may indicate that dry gas could also have accumulated in relatively deep regions, for example, the ramp or foredeep of the foreland basin. Traps under the faults could also be potential gas reservoirs.

As illustrated by this study in the Xuefeng uplift, the evolution of hydrocarbon systems can be extremely challenging to understand when affected by multiple tectonic events. This is particularly the situation in the Yangtze plate block, southern China (Mei et al., 2012; Yan et al., 2003). Although many hydrocarbon shows (e.g., bitumen, oil, and gas) are known (e.g., Weng'an, Nanshanpin, and Pingtang reservoirs; Deng et al., 2014), only the Sichuan Basin currently has producing fields (e.g., Puguang gas field; Ma et al., 2007). Furthermore, pyrobitumen occurs widely in basins all over the world, for example, the Alberta Basin (Canada), Dahoney basin (Nigeria), and Basque-Cantabrian Basin (Spain). Therefore, Re-Os bitumen and pyrobitumen geochronology (coupled with AFT dating) shows the potential to yield quantitative timing of oil and gas generation that may aid in the understanding of both the temporal and spatial evolution of hydrocarbon systems.

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