ORIGINAL PAPER-EXPLORATION GEOLOGY



Hydrocarbon source rock assessment of the shale and coal bearing horizons of the Early Paleocene Hangu Formation in Kala-Chitta Range, Northwest Pakistan

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Abstract

The present study aims to investigate the origin, type, thermal maturity and hydrocarbon generation potential of organic matter and paleo-depositional environment of the Early Paleocene (Danian) Hangu Formation outcropped in the Kala-Chitta Range of Northwest Pakistan, Eastern Tethys. Organic-rich shale and coal intervals were utilized for geochemical analyses including TOC (total organic carbon) and Rock–Eval pyrolysis coupled with carbon (δ^{13} Corg) and nitrogen (δ^{15} Norg) stable isotopes. The organic geochemical results showed that the kerogen Type II (oil/gas prone) and Type III (gas prone) dominate the investigated rock units. The TOC (wt%) and S_2 yield indicate that the rock unit quantifies sufficient organic matter (OM) to act as potential source rock. However, the thermal maturity T_{max} °C marks the over maturation of the OM, which may be possibly linked with the effect attained from nearby tectonically active Himalayan Foreland Fold-and-Thrust Belt system and associated metamorphosed sequences. The organic geochemical analyses deciphered indigenous nature of the OM and resultant hydrocarbons. The δ^{13} Corg and δ^{15} Norg stable isotopic signatures illustrated enrichment of the OM from both marine and terrestrial sources accumulated into the Hangu Formation. The Paleo-depositional model established using organic geochemical and stable isotopic data for the formation supports its deposition in a shallow marine proximal inner shelf environment with prevalence of sub-oxic to anoxic conditions, a scenario that could enhance the OM preservation. Overall, the formation holds promising coal and shale intervals in terms of organic richness, but due to relatively over thermal maturation, it cannot act as an effective source rock for liquid hydrocarbon generation and only minor amount of dry gas can be expected. In implication, the results of this study suggest least prospects of liquid hydrocarbon generation potential within Hangu Formation at studied sections.

Keywords Organic geochemistry · Stable isotopes · Organic matter · Paleocene coals · Thermal maturity

Introduction

The organic-rich shales and coals bearing stratigraphic sequences are widely distributed in Cenozoic and Mesozoic rock records (Petersen 2006; Friederich et al. 2016) and these sedimentary units generate and expel much of

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hydrocarbons in many parts of the world (Petersen et al. 2002; Pashin 2008; Ahmed et al. 2009; Hakimi et al. 2013; Sykes et al. 2014; Escobar et al. 2016; Chen et al. 2017; Khan et al. 2018). However, the heterogeneities in physical and chemical characteristics of such coals and shales are responsible for variation in oil/gas generation capacities (Abbassi et al. 2016), which is mostly constrained by depositional environment(s) and organic matter types of the rock units (Sykes 2001; Hakimi et al. 2013; Niu et al. 2019). Advancement in the petroleum industry and continuously rising energy demand have extensively attracted the attention of researchers towards the exploration of both conventional and unconventional hydrocarbon resources (Wang et al. 2019; Meng et al. 2020; Radwan et al. 2021a). The conventional hydrocarbon reservoirs are characterized by the migration of hydrocarbon from an external source rock into



the porous and permeable reservoir rock unit (Kassem et al. 2021; Radwan 2021; Radwan et al. 2021b; Shehata et al. 2021), whereas the unconventional hydrocarbon reservoirs are sharing the same source and reservoir rock unit (Radwan et al. 2020, 2021a). Although energy can be generated from various resources, but the most significant resource remains fossil fuels including coals and hydrocarbons (Domac et al. 2005; Askari and Krichene 2010). Furthermore, the socioeconomic development and long-term stability of certain countries are entirely dependent on their energy production from organic-rich shale and coal resources and its sustainable utilization (Askari and Krichene 2010; Malkani et al. 2016; Ullah et al. 2018). The hydrocarbon bearing, Tertiary coal and organic-rich shale deposits are reported worldwide for instance in the Gippsland Basin of Australia, Guasare Basin of Venezuelan, Columbus Basin of West Indies, Malaysia, Vietnam, Myanmar, China and Indus Basin of Pakistan (Boreham et al. 2003; Shah 2009; Alias et al. 2012; Hakimi et al. 2013; Abbassi et al. 2016; Escobar et al. 2016; Friederich et al. 2016; Khan et al. 2018). Such prospective rock units need detailed studies for hydrocarbon potential assessment using Rock-Eval pyrolysis, TOC analysis, isotopic proxies and organic geochemistry (Boreham et al. 2003; Hao et al. 2011; Makeen et al. 2015; Yandoka et al. 2015; Abbassi et al. 2016; Niu et al. 2019). Consequently, the investigation of coals and organic-rich shales as hydrocarbon resource is an attractive option to cope with rising energy demands and provide excellent research opportunities to tackle with associated organic geochemical and depositional complexities.

In this regard, the Early Paleocene Hangu Formation, which is one of the prospective formations, is targeted for its source rock potential evaluation in the Kala-Chitta Range of Northwest Pakistan (Fig. 1). Conventionally, this formation has acted as a proven resource of bituminous coal in the Upper Indus Basin of Pakistan (Kadri 1995; Ullah et al. 2018; Qureshi et al., 2020). However, in this study, it is assumed to act as a potential source rock for hydrocarbon generation to charge the associated reservoirs in the Kala-Chitta Range of Upper Indus Basin, Pakistan. The Hangu Formation represents a shallow marine stratigraphic unit of the eastern Tethyan affinity in Pakistan (Shah 2009; Craig et al. 2018; Qasim et al. 2020). Lithologically, it consists of light grey to reddish brown, fine to coarse-grained sandstone, medium to thick-bedded limestone with grey shale intercalations and coal beds (Shah 2009; Qureshi et al. 2020). It is widely distributed in Attock-Cherat, Kala-Chitta, Surghar and Salt ranges along with Kohat, Potwar and Hazara basins of Pakistan (Fig. 1; Shah 2001, Shah 2009; Malkani 2012; Malkani and Manhood 2016; Qureshi et al. 2020). It has been studied extensively by several researchers notably Ahmed et al. (1986), Shah et al. (1993) from stratigraphic, sedimentological, provenance and economic point of view. Ahmed et al. (1986) suggested that the Hangu Formation



represents accumulation of clastic facies of the Paleocene age and is a host rock for coal and organic-rich shale intervals. The coal occurs in the form of lenses and beds in the upper argillaceous facies of the formation (Figs. 2 and 3; Hussain et al. 1990; Shah 2009; Latif et al. 2021). Qasim et al. (2020) worked out its petrography and geochemistry and concluded that the sediments of the Hangu Formation preserve global Cretaceous-Tertiary unconformity (i.e. KT-boundary). Based on spinel geochemistry and U-Pb dating, it is suggested that the sediments of the Hangu Formation are primarily derived from the Tethyan Himalayan source area (i.e. Indian Plate; Qasim et al. 2020). Shah (2001) described the lithofacies including (1) Lateritic/Bauxitic lithofacies, (2) Cross-bedded sandstone lithofacies, (3) Bioclastic limestone lithofacies, (4) Coal and carbonaceous shale lithofacies, and (5) Bioturbated quartzose sandstone lithofacies (Fig. 2). Among these lithofacies the "Coal and carbonaceous shale lithofacies" holds importance from the perspective of hydrocarbon exploration and production. The characterization and investigation of lithofacies and depositional environment for coal beds help to predict variability in coal bed's physical and chemical characteristics. Likewise, the investigation of coal-bearing paleo-environments has regional and worldwide applications for hydrocarbon resource exploration and exploitation (Warwick and Shakoor 1993; Hakimi et al. 2013; Ullah et al. 2018). Despite of the aforementioned studies, detailed investigations regarding organic geochemical characteristics, thermal maturity and source rock potential of the Hangu Formation at Kala-Chitta Range relatively remain insufficient and need detailed studies to address the desired gap in knowledge.

In this regard, this study is focused to investigate the origin, types, organic geochemical characteristics and thermal maturity of the organic-rich shale and coal intervals encountered within the Hangu Formation at Kala-Chitta Range, Northwest Pakistan. Additionally, the hydrocarbon generation potential of the formation is evaluated and its source rock potential is assessed. A paleo-depositional model is established to understand the paleo-depositional constrains on organic matter (OM) preservation within Hangu Formation. Kerogen types encountered within the formation are identified, and stable isotopic proxies are used to illustrate origin and depositional conditions of the OM. In implication, the results of this study hold importance to contribute considerably to the source rock quality assessment of the Hangu Formation during exploration and production campaigns in the Kala-Chitta Range of Northwest Pakistan.

Geologic setting and stratigraphic evolution

Pakistan overlaps a region, where three active plates (i.e. Indian, Arabian and Eurasian plates), converge and interact with each other and has greatly re-shaped its tectonic and

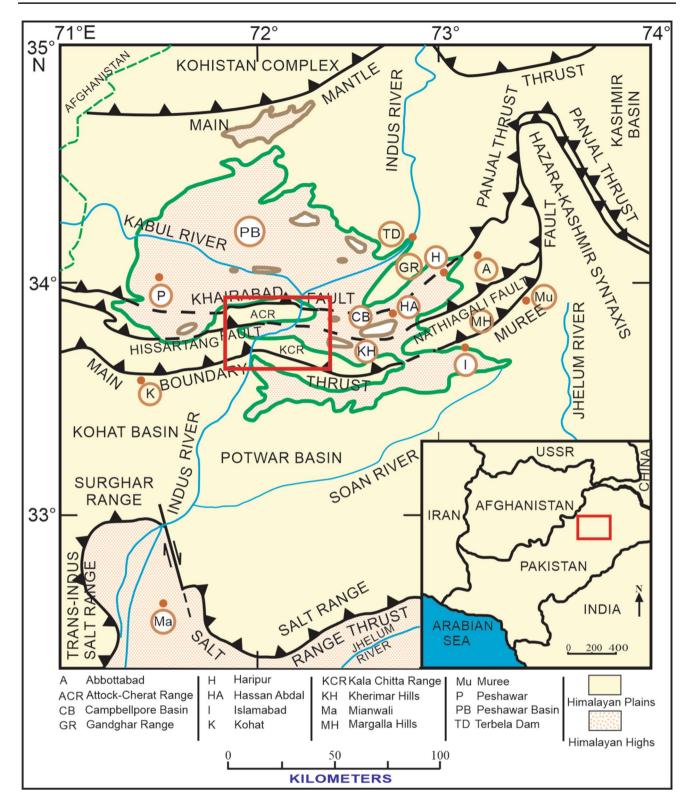


Fig. 1 Tectonic map of North Pakistan, showing location and tectonic setting of the study area (Red rectangle) along with major thrust faults and rivers. Map inset shows location of the study area with respect to Pakistan and surrounding countries (After Hylland et al. 1988)



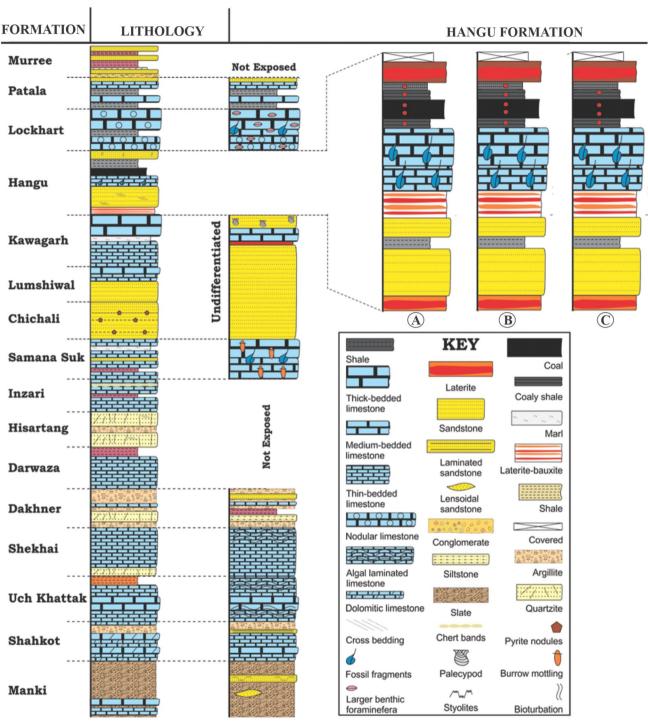


Fig. 2 Stratigraphic column illustrating general stratigraphy of the study area (left side) and lithologic logs (right upper side) of the Hangu Formation as studied sections (Adapted from Shah 2001;

Latif et al. 2021). (A) Khairabad section (B) Gandab section and (C) Kahi section. The logs are not to the scale as these are adapted from Latif et al. (2021)

structural framework (Kazmi and Jan 1997). The collision between Indian and Eurasian plates resulted into various thrust systems including Main Karakorum Thrust (MKT), Main Mantle Thrust (MMT), Main Boundary Thrust (MBT) and Salt Range Thrust (SRT) from north towards south of Pakistan (Fig. 1; Kazmi and Jan 1997; Acharyya 2007; Ullah et al. 2020a). Further northward drift has resulted into various small-scale thrust and normal faults



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Fig. 3 Field illustrations of the Hangu Formation at studied sections A Upper contact of the Early Paleocene Hangu Formation with overlying Middle Paleocene Lockhart Limestone B Faulted contact between Manki Formation and Lockhart Limestone C Organic-rich shale having fissility in upper part of the Hangu Formation D Lower

contact of the Hangu Formation with under lying Kawagarh Formation along with associated laterites **E** Carbonaceous shale of the Hangu Formation having coal patches **F** Abandoned coalmine targeting the coal horizons of the Hungu Formation at Khairabad section

and associated splays, which have subsequently caused a complex local geology of the study area (Fig. 1; Hylland et al. 1988; Yaseen et al. 2021). The study area (Kala-Chitta Range) is an integral part of north-western margin of the Indian Plate which emerged as a consequence of successive uplifting along the MBT thrust about 1.9 million years ago (Burbank and Raynolds 1988; Ullah et al. 2020a). The Kala-Chitta Range (KCR) is bounded by Attock-Cherat Range in the north and Potwar Basin in the south, whereas Margalla Hills are located in the northeast and Kohat Basin in the southwest (Fig. 1; Burbank et al. 1989; Ullah et al.

2020b). The studied rock unit (i.e. Hangu Formation) of Khairabad, Gandab and Kahi sections is stratigraphically an integral part of the Kala-Chitta Range (Figs. 1 and 2; Shah 2009; Yeats and Lawrence 1982). Tectonically, the Kala-Chitta Range is making the northern periphery of the Potwar Basin and represents part of the Lesser Himalayas (Yeats and Hussein 1987). Geologically, the Kala-Chitta Range is highly deformed and contains thrust faults, anticlines, pop up structures and hill ranges (Figs. 2; Kazmi and Jan 1997; Shah 2009; Awais et al. 2013). The Hangu Formation of the Khairabad section is stratigraphically, an



integral part of the Kala-Chitta Range, however, due to back thrusting phenomena its slices are located in Attock-Cherat Range as well. The Kala-Chitta Range, located in Northwest Pakistan, is the most intriguing locality exposing the Triassic to Recent sediments. The Gandab and Kahi sections are located in Kala-Chitta Range, which constitute part of Hill Ranges of northern Pakistan (Yeats and Lawrence 1982). The Nizampur Basin is located in the northwestern side of the Kala-Chitta Range, north of the Main Boundary Thrust (Awais et al. 2013). The Kala-Chitta Range is a narrow strip of the mountainous belt, bound by the Hazara Mountain in the east and Samana Range in the west, Khair-e-Murat Hills and Gandghar Ranges are located to the northeast, while MBT marks its southern boundary (Fig. 1; Yeats and Lawrence 1982; Yaseen et al. 2021). Age of uplifting of the Kala-Chitta Range along MBT is approximately 1.9 million years, whereas the Attock-Cherat Range has been uplifted about 3 million years ago (Burbank et al. 1989). The stratigraphic succession exposed in the study area includes the Khairabad, Gandab and Kahi sections and is shown in Fig. 2. The Paleocene Succession exposed in the study area is comprised of Makarwal Group that include the Early Paleocene Hangu, Middle Paleocene Lockhart and Late Paleocene Patala formations (Figs. 1 and 2). The former formation is important from the perspective of hydrocarbon source assessment, whereby hosting the organic-rich shale and coal intervals.

Material and methods

The research protocol adopted for this study includes field sampling and laboratory analyses.

Field investigation and sampling

During field campaign, the study sections including Khairabaid, Gandab and Kahi occurring in the Kala-Chitta Range of Northwest Pakistan were measured, photographed and sampled (Figs. 1, 2 and 3). Diagnostic field features were noted and illustrated on the lithological logs (Fig. 2). A sum of fourteen rock samples from fresh organic-rich outcrops and coalmines were collected for laboratory analyses. The samples were packed into polythene bags, marked with appropriate sample number and locality, stored at low temperature approximately (10 °C) to reduce microbial degradation and oxidation of the OM. The rock samples were washed gently with distilled water for removal of contamination and exogenous material. The samples were then dried, powdered and consequently subjected for TOC analysis and Rock-Eval pyrolysis at the laboratories of Hydrocarbon Development Institute of Pakistan (HDIP), Islamabad,



Pakistan. Details about sampled sections and rock units are given below.

Khairabad section

At Khairabad Sect. (33° 51.835 N, 072° 13.975 E), the Hangu Formation is mainly comprised of fine to medium grained compacted sandstone, carbonaceous shale, coal and laterite beds (Fig. 3A–B). The shale is organic-rich, fissile and is characterized by thin lamination. The coal beds (3 m) are grey to black and contain sulphur stains and rusting due to surface weathering. The coal beds are in sharp contacts with the shale beds in upper part of the Hangu Formation. The coal also possesses pyritic nodules and rarely calcite veins at places. The coals and shales are dominated in upper part of the formation. The lower contact of the formation in not exposed, whereas the upper contact is conformable with overlying Middle Paleocene Lockhart Limestone (Fig. 3B).

Gandab section

The Hangu Formation consists of laterite, carbonaceous shale and coal beds along with thin beds of limestone (Fig. 3D–E) at Gandab Sect. (33° 46.434 N, 072° 09.882 E). At this locality, the Hangu Formation is comprised of light-grey, fine-grained, siltstone, dark-grey shale and minor carbonaceous shale having coal patches. The sandstone is calcareous, burrowed, flat or ripple bedded and interbedded mostly with siltstones. Although, coal mines are present in this area but are mostly abandoned due to extensive water logging and associated extraction problems. The Hangu Formation in the Gandab section unconformably underlies the Late Cretaceous Kawagarh Formation and conformably overlies the Middle Paleocene Lockhart Limestone.

Kahi section

At Kahi Sect. (33° 48.676 N, 072° 03.879 E), the Hangu Formation is not exposed at surface, however, the data presented were collected from coalmines (Fig. 3E). In the coalmines, the thick coal interval is present along with organic-rich shale intervals. The coalmines are strip mines, which targeted the coal intervals of the Hangu Formation for extraction. The coal bed intervals encountered within the coalmines are tentatively plotted on the lithologic logs to illustrate the lateral facies continuity within Hangu Formation (Figs. 2 and 3). The coal beds have sharp contacts with organic-rich shales. The Hangu Formation also contains reddish-brown laterites in upper and lower part of the succession.

Laboratory analyses

Organic geochemical analyses including TOC (wt%) and Rock–Eval pyrolysis were applied on the representative rock samples for source rock characterization and hydrocarbon potential evaluation (Table 1). These analyses were performed in the Laboratories of Hydrocarbon Development Institute of Pakistan (HDIP), Islamabad, Pakistan.

TOC analysis

TOC analysis involves heating of 100 mg pulverized rock sample at constant computer-programmed temperature in the helium inert environment, which gives TOC values in weight percent (wt%). For TOC analysis of the representative rock samples, the TOC analyzer (LECO-CS 300) was used. TOC measurement provides the preliminary insights about the organic richness of a source rock (Hunt 1996; De Souza et al. 2021). The rock samples with TOC (wt%) values higher than 0.5% were subjected to further organic geochemical analysis (i.e. Rock–Eval pyrolysis).

Rock Eval-pyrolysis

The Rock-Eval analysis was undertaken using a Rock-Eval-6 analyzer (Vinci Technologies) in accordance to the standard procedure proposed by Behar et al. (2001). The Rock-Eval pyrolysis parameters including S₁ (free hydrocarbon) S₂ (cracked hydrocarbon or generative potential), S₃ (expulsion of oxygen containing compound) yields, hydrogen index (HI), oxygen index (OI), production index (PI), and Temperature of maximum T_{max} (°C) are calculated and compared with published source rock standards (e.g. Peters 1986; Hunt 1996). The T_{max} (°C) is used as thermal maturity indicator to estimate the maturation levels of the organic-rich horizons within the formation. The HI and OI are helpful in understanding the quality of the OM. These parameters are complementary in investigating kerogen types, quality and hydrocarbon generation potential of source rocks (Hunt 1996; Behar et al. 2001; El Diasty et al. 2017; Hakimi et al. 2020). The Rock-Eval pyrolysis was performed on the selected rock samples from Hangu Formation at HDIP, Islamabad, Pakistan.

Stable isotopic analysis

The organic geochemical analyses were subsequently supplemented with stable isotopic proxies to attain more insights about the origin, types and depositional environment(s) of the OM encountered within the Hangu Formation. For stable isotope analysis, the powdered rock samples were measured (10 mg) with Sartorius MC-5 (electronic balance) and consequently, loaded into silver cups for decarbonisation with 5% hydrochloric acid (HCl). Subsequently, the neutralised samples were subjected into an elemental analyser connected with isotope ratio mass spectrometer (EA-IRMS) for C and N contents and their stable isotopic ratio was calculated. The elemental analyzer (Thermo Flash 2000 HT) and isotope ratio mass spectrometer (IRMS-Thermo DELTA V Advantage) were maintained at 1020 °C for analysis (Uveges et al. 2019). In EA-IRMS, the samples were combusted and resultant gases were separated into respective columns detected by the thermal conductivity detector. The gases (CO₂ and N_2) were consequently reduced to their respective elements, and the quantities were measured in parts per million ($\%_o$). V-PDB (Vienna Pee Dee Belemnite) was used as reference for C isotopes and natural air was used as standard for N isotopes (Gross et al. 2015; El-Diasty et al. 2017; Uveges et al. 2019). In this study, the δ^{13} C and δ^{15} N stable isotopes analyses were performed to interpret the origin, composition and types of OM as well as depositional conditions of the Hangu Formation. Carbon and nitrogen stable isotopes ratios (C/N) are used to distinguish various types of OM such as marine and terrigenous OM (Geel et al. 2015; El-Diasty et al. 2017). These analyses were performed at the Department of Earth and Environmental Sciences, KU Leuven, Belgium.

Results and discussion

TOC results

TOC values of the samples analysed, vary between 0.09 wt% (HAM-1C) and 61.43 wt% (HKM-4A) with an average value of 26.03 wt% (Table 1 and Fig. 4). The coal samples HKM-4A and HAM-1A contain the highest measured TOC (61.43) and (40.98) wt%, respectively, whereas the organicrich shale samples HAM-1C and HGM-3A show the lowest TOC (0.09 and 0.69) wt%. The higher TOC values are indicative of organic richness of the analysed beds and display moderate to good potential for the studied source rock. In the current data set, most of the samples exhibited TOC value higher than 2%, while the minimum TOC limit for a potential source rock is 0.5 wt% (Peters 1986; Hunt 1996), which implies promising TOC contents within the targeted intervals of the Hangu Formation. These higher TOC values show higher organic enrichment within the analysed rock samples probably under reducing or anoxic paleo-environmental conditions during deposition of the Hangu Formation, which subsequently enhanced the OM preservation (Khan et al. 2018; Latif et al. 2021). The higher TOC values are making the coals beds comparatively promising intervals but subject to other parameters such as Rock-Eval pyrolysis and thermal maturity. In terms of TOC results, it can be inferred that the Hangu Formation holds moderate to good source rock quality at studied sections (Table 1). Although



Sections	Sample No	TOC (wt%)	S ₁ mg HC/g rock	S ₂ mg HC/g rock	S ₃ mg CO ₂ /g rock	GP mg HC	HI mg HC/g TOC	OI mg CO ₂ /g TOC	PI mg HC	$^{T}_{ m CC}$
Gandaband and Kahi sections	HKM-4B	I	I	1	I	1	I	I	I	I
	HKM-4A	61.43	0.59	40.71	3.97	41.30	66.00	7.00	0.01°	480
	HGM-3D	I	I	I	I	I	I	I	I	I
	HGM-3C	15.96	0.13	5.54	4.40	5.67	35.00	28.00	0.02	462
	HGM-3B	I	I	I	I	I	I	I	I	I
	HGM-3A	0.69	0.02	0.13	0.47	0.15	19.00	68.00	0.13	516
Average	II	26.03	0.25	15.46	2.95	15.71	40.00	34.33	0.08	486
Khairabad `section	HAM-2D	I	I	I	I	I	I	I	I	I
	HAM-2C	2.43	0.03	0.06	0.07	0.09	3.00	3.00	0.33	525
	HAM-2B	I	I	I	I	I	I	I	I	I
	HAM-2A	2.16	0.02	1.37	0.98	1.46	63.00	45.00	0.01	527
	HAM-1D	I	I	I	I	I	I	ļ	I	I
	HAM-1C	0.09	0.01	0.08	0.06	0.09	89.00	67.00	0.11	480
	HAM-1B	I	I	I	I	I	I	I	I	I
	HAM-1A	40.98	0.04	1.42	0.97	1.46	4.00	2.00	0.03	528
Average	II	11.42	0.03	0.73	0.52	0.78	39.75	29.25	0.12	515

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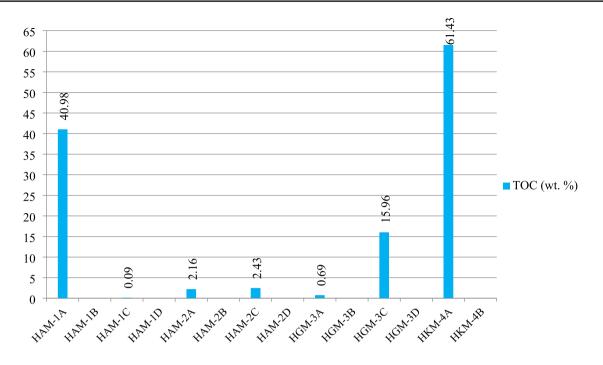


Fig. 4 Graphical representation showing organic matter enrichment of the rock samples of the Hangu Formation

TOC is a good indication of organic richness of a formation, however, TOC alone can seldom evaluate a source rock precisely. Thus, additional insights from Rock–Eval pyrolysis and other essential parameters (e.g. HI, OI and T_{max} °C) are needed in order to assess the source rock potential appropriately (Hakimi and Abdullah 2014; Khan et al. 2021).

Rock–Eval pyrolysis results

The Rock-Eval pyrolysis results of the representative rock samples are given in Table 1 and Fig. 5. The results of S_1 yields (free hydrocarbon) range from 0.01 mg of HC/g rock (HAM-1C) to 0.59 mg HC/g rock (HKM-4A) with an average of 0.11 mg HC/g rock at studied sections. Similarly, the S₂ yields range from 0.06 (HAM-2C) to 40.71 (HKM-4A) with an average value of 15.46 mg HC/g rock. These ranges in S_1 and S_2 yields demonstrate the source rock quality that ranges from moderate to good quality. The values of relevant parameters of Rock-Eval pyrolysis such as S₃ yields, hydrogen index (HI), production index (PI), genetic potential (GP), oxygen index (OI), and thermal maturity are measured and provided in Table 1. The average GP while accumulating both the free hydrocarbon (S_1) and cracked hydrocarbon (S_2) is 15.71 and 1.44 (mg HC), respectively, as studied sections. The hydrogen index (HI) is ranging from 3 mg HC/g TOC (HAM2-C) to 89 mg HC/g TOC (HAM-1C) with an average of 39.75 mg HC/g TOC (Table 1). Likewise, the average OI is $34.13 \text{ (mg CO}_2/\text{g TOC)}$ and $29.25 \text{ (mg CO}_2/\text{g TOC)}$ at studied sections. The relatively lower HI (<100) and OI (< 50) demonstrate derivation of the OM mainly from terrestrial sources (Hunt 1996; Hakimi et al. 2013). The lower OI within coal samples also designates a good quality coal with high heat capacity (Latif et al. 2021). The Rock–Eval $T_{\rm max}$ is lying between 462 and 528 °C with an average of 515 °C displaying an increasing trend from carbonaceous shales samples to coals (Table 1). Considering the Peters (1986) and Hunt (1996) guidelines, the rock samples having S₂ values ranging between 5 and 10 mg HC/g rock are considered to have good source rock quality, whereas those holding S₂ values less than 5 mg HC/g are classified as poor source rock potential. In case of the Hangu Formation, the rock samples having S₂ yield higher than (> 5 mg HC/g rock) are considered good quality source rocks intervals.

Stable isotopic proxies

Carbon (δ^{13} Corg) and nitrogen (δ^{15} Norg) stable isotopic results are presented in Table 2. The δ^{13} Corg values vary between – 22.2 and – 26.2‰ with an average of – 24.43‰, whereas the δ^{15} N signatures range from 4.10 to 4.60‰ with an average of 4.25‰ (Table 2). Likewise, the C/N elemental ratio ranges from 1.45 to 14.02 with a cumulative value of 7.67 and the nitrogen contents are ranging from 0.16 to 0.35% with an average of 0.28% (Table 2). The negative value of δ^{13} Corg above (> – 24‰) shows the derivation of the OM from terrestrial organic materials, whereas the value below (< – 24‰) illustrates the derivation of OM from marine sources (Könitzer et al. 2016; El



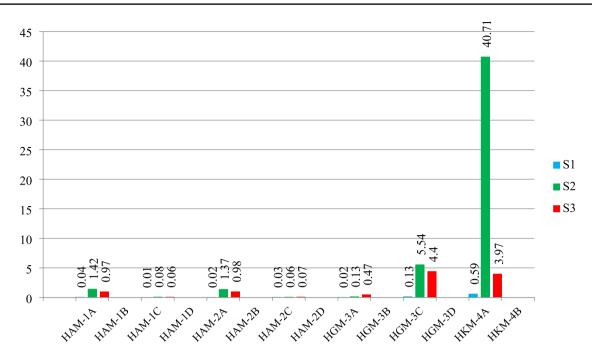


Fig. 5 Histogram illustrating the Rock–Eval pyrolysis S_1 , S_2 and S_3 yields of the analysed rock samples of the Hangu Formation

Table 2 TOC, TON, C/N elemental ratio and stable	Sample No	TOC (wt%)	C (%)	N (%)	C/N	δ^{13} Corg (‰)	δ^{15} Norg (‰)
isotope signatures of rock	HKM-4B	_	_	_	_	_	_
isotope signatures of rock samples of the Hangu Formation	HKM-4A	61.43	32.40	0.18	1.45	-22.20	4.20
Formation	HGM-3B	_	-	-	-	_	_
	HGM-3A	0.69	0.42	0.41	9.00	-26.20	4.10
	HAM-2B	-	-	-	-	_	_
	HAM-2A	02.16	02.06	0.35	6.20	-25.10	4.60
	HAM-1B	-	-	-	_	_	_
	HAM-1A	40.98	22.6	0.16	14.02	-24.20	4.10
	Average =	26.32	14.37	0.28	7.67	-24.43	4.25

Diasty et al. 2017). Likewise, the higher C/N ratio illustrated the dominance of OM of terrigenous origin, whereas lower values (< 10) show derivation of the OM from marine sources (Geel et al. 2015). In general, the ratio of terrigenous and marine OM is a function of OM influx and provides essential information about origin of the organic matter (Uveges et al. 2019). Additionally, the more negative carbon (-26.2%) indicated marine OM possibly from phytoplanktons and algae (Geel et al. 2015; Uveges et al. 2019). The overall isotopic proxies portrayed that Hangu Formation was deposited in a proximal inner shelf shallow marine depositional setting in association with terrestrial environment with influx of tremendous OM from terrestrial environment and minor amount from marine origin. The preservation of OM within Hangu Formation is possibly because of the development of the sub-oxic to



anoxic conditions most likely due to rapid consumption of oxygen by micro-organism in shallow marine settings and plenty of organic influx. In some of the samples, the carbon seems to be derived from wood fragments as its values ranges from -22.2 to -22.4%, especially in coal samples. The higher TOC and dark black colour of coal and shale beds reveals the oxygen deficient conditions prevailed during coal and organic-rich shale deposition, which consequently boosted the OM preservation. Generally, fluctuation in sediment supply, bio-productivity and dilution by siliciclastics influenced the burial rate and preservation of OM (Killops and Killops 2005; Naeher et al. 2019), however, in case of Hangu Formation the organic enrichment is relatively higher in coal and organic-rich shales that is constrained by sub-oxic to anoxic conditions and plenty of OM influx in the formation.

Source rock characteristics and hydrocarbon potential evaluation

Organic matter (OM) types, quantity and thermal maturity are important considerations used in the evaluation of both conventional and unconventional hydrocarbon systems (Abbassi et al. 2016). The source rock parameters used to assess the hydrocarbon generation potential of the Hangu Formation included;

Source rock generative potential

The source rock generative potential of Hangu Formation is assessed using TOC (wt%) versus S_2 yield, genetic potential ($S_1 + S_2$) versus TOC, HI versus TOC and S_1 versus TOC (Fig. 6A-D), respectively (Hakimi et al. 2013; Tahoun and Deaf 2016; El-Khadragy et al. 2018). Generally, the TOC values lie in the range of 1–100 (wt%) and S_2 values lie in the range of 0.1–1000 (mg HC/g rock) for all organic-rich lithologies in sedimentary basins (Hunt 1996; Hakimi and

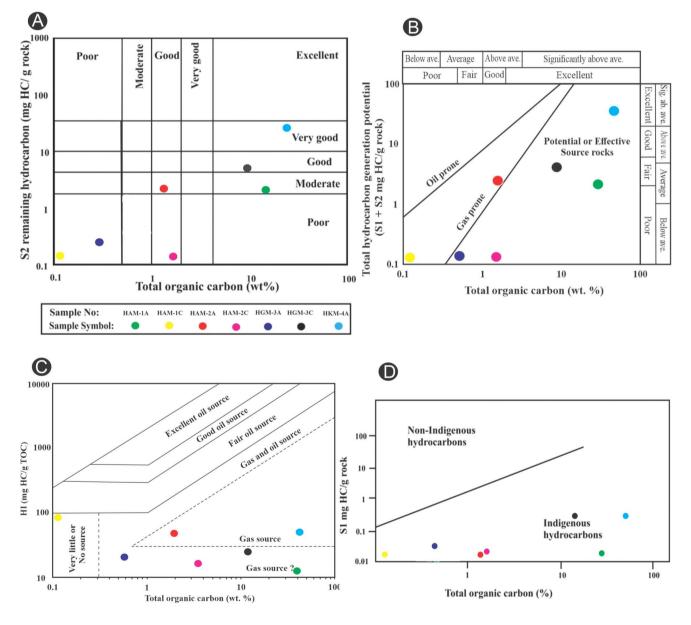


Fig. 6 Source rock plots illustrating organic geochemical characteristics of the Hangu Formation \mathbf{A} S₂ versus TOC plot illustrating types and quality of the source rock encountered within Hangu Formation (After Hakimi and Abdullah 2014) **B** Genetic potential versus TOC indicating expected hydrocarbon types and quality of the source rocks

(After El Nady et al. 2015) **C** Generating potentialities of source rocks of the Hangu Formation based on HI and TOC (After De Souza et al. 2021) **D** S₁ versus TOC plot deciphering indigenous nature of the hydrocarbon encountered within the Hangu Formation (After Khan et al. 2018; De Souza et al. 2021)



Abdullah 2014), however, in case of the Hangu Formation, the S_2 yield ranges from 0.06 mg HC/g rock (HAM-2C) to 40.71 mg HC/g rock (HKM-4A) showing poor to very good source rock potential (Table 1). Thus, TOC values of 0.09–61.43 (wt%) and S_2 values of 0.06–40.71 mg of HC/g rock are indicating poor to very good and excellent source rock quality, respectively (Hunt 1996; El Diasty et al. 2017; Hakimi et al. 2020; Khan et al. 2021). Relationship between the genetic potential (GP) and TOC shows that the studied formation is considered as poor to excellent source potential (Fig. 6B). On the other hand, the plot of TOC versus HI shows oil and gas potential except for HAM-1A, which shows a barren or non-source rock characteristics (Fig. 6C). The black shiny coal (HKM-4A) holds considerable hydrocarbon generation potential at the studied sections (Table 1). However, the grey shale (HAM-1C) acts as poor to fair source rock potential (Fig. 6A-D). Consequently, most of the samples contain above average levels of the organic matter (TOC > 0.5 wt%) and these values meet the accepted standards of a source with poor to very good and excellent hydrocarbon generative potential (Hunt 1996; Hakimi and Abdullah 2014). The S_1 versus TOC diagram is used to discriminate between indigenous and non-indigenous hydrocarbons (Fig. 6D; El-Khadragy et al. 2018). All studied rock samples have relatively low S1 value and relatively high TOC contents, which indicated that the analysed rock samples are free from contamination or exogenous material and the hydrocarbon is of indigenous origin (Khan et al. 2021).

Kerogen types and organic matter

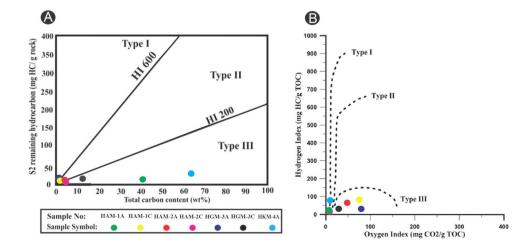
Essential information on the types of kerogen (organic matter) is attained from relevant parameters of Rock–Eval pyrolysis such as S_2 yield, HI and OI supplemented with TOC analysis (e.g. Peters 1986; Hunt 1996). It reveals the quality or nature of OM to assess the hydrocarbon generating potential of a source rock (Makky et al. 2014; Hakimi et al. 2020). Different types of kerogen can produce different types of hydrocarbons (Hunt 1996; Hakimi and Abdullah 2014; Hakimi et al. 2020). It is therefore necessary to identify the quality of OM encountered within the formation. Different plots between HI versus TOC, HI versus OI and HI versus T_{max} (°C) are used to identify kerogen type (Fig. 7A–B; Mustapha and Abdullah 2013; Hakimi and Abdullah 2014; Abbassi et al. 2016). The HI lies in the range of 3 (HAM-2C) to 89 (HAM-1C) and TOC ranges from 0.09 (HAM-1C) to 61.40 (HAM-4A), while the OI ranges between 2 (HAM-1A) and 68 (HGM-3A) (mg CO_2/g TOC) shown on the modified van Krevelen's diagram (Fig. 7A-B) which confirmed that the analysed samples from Hangu Formation fall in the zone of gas prone Type III kerogen and minor oil/gas prone Type II kerogen. However, the OM as inferred from organic geochemical proxies is primarily dominated by terrestrial Types III kerogen compared to marine Type II kerogen which indicates plenty of terrestrial OM (Fig. 7A-B). The TOC content in a source rock determines the quantity of the generated hydrocarbons, whereas the kerogen types constrain the type of the generated hydrocarbons (Boreham et al. 2003; Abbassi et al. 2016). Despite of relatively low HI values, some of the coals are interpreted to have capability of generating gaseous hydrocarbons within the Hangu Formation. Unlike the organic-rich shales, the coal beds of the Hungu Formation are dominated by terrigenous OM and hold over-mature kerogen Type III (Fig. 7).

Thermal maturity level

Thermal maturity estimation is essential to know about the quality of a source rock (Peters 1986). T_{max} (°C) temperature is considered as one of the most reliable indicators of thermal maturation of the OM in sediments and can be affected by burial and the kerogen types (Hunt 1996; Tahoun and Deaf 2016). The Rock–Eval pyrolysis T_{max} (°C) provided direct insights about the OM thermal maturity of the

Fig. 7 Modified van Krevelen's diagrams illustrating kerogen types of source established from organic geochemical parameters A Rock–Eval pyrolysis S2 versus TOC displaying kerogen types encountered within Hangu Formation (After Hakimi and Abdullah 2014) B Modified van Krevelen's diagram showing organic matter types based on HI and OI encountered within the Hangu Formation (After; Khan et al. 2018; De Souza et al. 2021)





source rock intervals of the Hangu Formation (Table 1). The achievement of optimum thermal maturity is essential, as kerogen Type I is regarded mature at T_{max} of 435–450 °C, kerogen Type II is mature between 420 and 460 °C and kerogen Type III becomes mature at a temperature range of 400–500 °C (Hakimi and Abdullah 2014; El-Khadragy et al. 2018). The immature or post mature source rocks are not capable of generating liquid hydrocarbons except minor amounts of dry gas (Khan et al. 2021). The assessment of thermal maturity of OM in the Hangu Formation was carried out using plots, HI versus T_{max} (Fig. 8). HI values lie in the range of 3–89 mg HC/g TOC, whereas T_{max} values range from 462 °C (HGM-3B) to 528 °C (HAM-1A) with an average value of 515 °C as shown in Table 1. The Rock–Eval $T_{\rm max}$ values and HI of the studied samples illustrated variation in thermal maturity of the analysed samples that is largely constrained by burial history and OM types of the formation (Fig. 8; Khan et al. 2021). The term thermal maturation is widely accepted to describe the degree of diagenetic evolution of the OM and its alteration into generation of hydrocarbon which is mostly estimated through measurements of T_{max} (°C) (Hunt 1996; Latif et al. 2021). The higher T_{max} shows that the organic rich sediments of the Hangu Formation are thermally over-mature for liquid

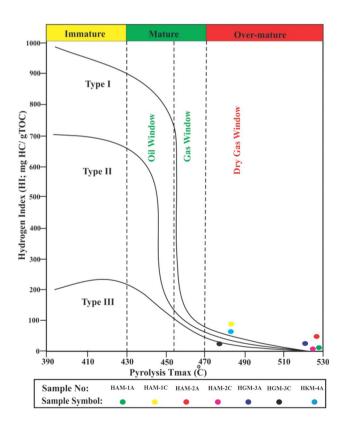


Fig.8 Plot of HI versus Rock–Eval pyrolysis- T_{max} indicating kerogen types and thermal maturity levels of the analyzed rock samples of the Hangu Formation (After Khan et al. 2018; De Souza et al. 2021)

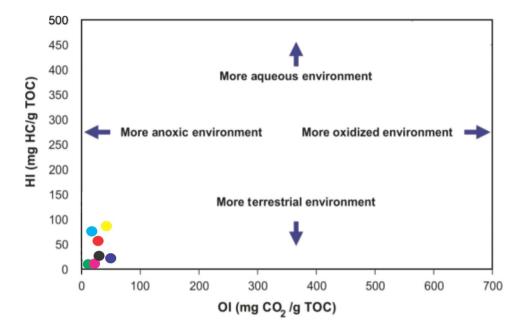
hydrocarbon generation but can possibly act as a source rock for dry gas generation (Khan et al. 2018). The possible reason for this high thermal maturity of Hangu Formation in the Kala-Chitta Range is due to its proximity of the nearby tectonically active Himalayan Foreland Fold-and-Thrust Belt system and associated metamorphosed sequences because of the Himalayan orogeny (Shah 2001; Latif et al. 2021). These processes enhance the coalification of OM, which in turn attain higher thermal maturities (Latif et al. 2021).

Depositional setting

The characterization and investigation of organic-shale and coal-bearing paleo-environment(s) has regional and worldwide applications in the exploration of fossil fuels resources (Warwick and Shakoor 1993; Hao et al. 2011; Ullah et al. 2018). Additionally, the interpretation of depositional environment for coal and organic rich shales helps to predict variability in its physical and chemical characteristics that ultimately constrains its source rock potential (Hakimi et al. 2013; Friederich et al. 2016). In light of available literature of Shah (2001), Shah (2009) and Oureshi et al. (2020) detail sedimentological studies of Hangu Formation indicated that it represents a wide variation in lithology encompassing sandstone, siltstone, limestone, coal and shale intervals. Five distinct lithofacies are distinguished within the Early Paleocene Hangu Formation that include (1) Lateritic/Bauxitic lithofacies, (2) Cross-bedding sandstone lithofacies, (3) Bioclastic limestone lithofacies, (4) Coal, carbonaceous shale lithofacies and (5) Bioturbated quartzose sandstone lithofacies. Out of these the coal- and carbonaceous shale-lithofacies are assessed, that possess suitability to act as source rock intervals for hydrocarbon generation. In this study, the term organic-rich shale is used for carbonaceous shale lithofacies. The modified van Krevelen (1993) diagrams have been considred in the current study for the interpretation of depositional enviornment(s) of the organic-rich intervals within the studied formation (Fig. 9). The investigated coal and shale samples indicated terrestrial to proximal inner shelf shallow marine depositional setting with prevalence of sub-oxic to anoxic depositional conditions (Figs. 9 and 10). The dark black colour of shale units and presence of pyrites also demonstrates prevalence of sub-oxic to anoxic conditions (Killops and Killops 2005). The organic geochemical proxies including HI and OI lower values show that the OM is mainly derived from terrestrial sources with minor marine influx (Khan et al. 2021; Latif et al. 2021). The identified kerogen Type II and Type III also indicate a mixed proximal inner shelf shallow marine environment with a terrestrial swampy sub-environment. The preservation of OM is enhanced due to existence of sub-oxic to anoxic conditions that established possibly because of the rapid oxygen consumption by micro-organism and plenty of organic



Fig. 9 Modified van Krevelen's diagram elucidating origin and depositional environment of the organic matter encountered within Hangu Formation in Kala-Chitta Range (After Last and Ginn 2005)



influx into shallow marine settings (Hunt 1996; Naeher et al. 2019). The carbon (δ^{13} Corg) and δ^{15} Norg signatures along with C/N elemental ratio also show the derivation of OM mainly from terrestrial sources as well as minor marine inputs. Ratio of terrigenous and marine OM is a function of organic matter influx that indicates that OM is derived from both sources (Tyson 1995; Uveges et al. 2019). The stable isotopic proxies also showed that anoxic conditions prevailed during coal and organic-rich shale deposition that enhanced preservation of the OM as indicated by extremely negative values of δ^{13} Corg (Table 2). The overall isotopic proxies portrayed that the Hangu Formation was deposited in a shallow marine proximal inner shelf setting in close proximity to terrestrial environment, which attained tremendous influx of OM from terrigenous sources and minor amount from marine sources (Fig. 9).

Origin, abundance and preservation of organic matter

Relevant factors influencing source rock quality include the primary productivity, OM input, thermal maturation and preservation conditions of the OM within a depositional sequence (Tyson 1995; Alias et al. 2012; Naeher et al. 2019). The TOC (wt%), Rock–Eval pyrolysis S_1 , S_2 and HI values are providing insights about the abundance of OM matter a feature which supports source rock potential of the formation. The higher TOC values suggest that the Hangu Formation is enriched in OM and possess suitability for source rock prospects (Table 1). Since the formation is deposited in a shallow marine proximal inner shelf setting (Fig. 10), where the redox conditions are considered essential for OM preservation as anoxic water is more beneficial for OM



preservation than oxic conditions (Tyson 1995; Makeen et al. 2015). Generally, in shallow marine proximal inner shelf setting source rocks, the OM abundance ranges from < 1 wt%to > 20 wt% TOCs with varying kerogen types from Type II to Type III, whereas deeper marine sediments are characterized by Type I kerogen (Peters 1986; Abbassi et al. 2016). Stable isotopic signatures elucidated that the Hangu Formation holds mixed organic matter, which is sourced from both marine and terrestrial environment(s). Additionally, the marine OM is predominantly derived from phytoplanktons, bacteria, fungi and microorganisms, whereas the terrestrial OM is derived from humic material of higher plants. The presence of negative carbon in the organic-rich shales suggests reducing conditions during deposition owing to the plenty of nutrients and contribute to good preservation of OM (Makeen et al. 2015; Uveges et al. 2019). Carbon and nitrogen stable isotopes ratios $(\delta^{13}C_{org})$ distinguished various types of OM such as marine and terrigenous OM (Geel et al. 2015; El-Diasty et al. 2017). Carbon stable isotopes also illustrated differences in organic influx and depositional conditions. It is also inferred that the OM enriched in isotopically light carbon isotopes indicates deposition in suboxic to anoxic depositional conditions (Shah 2001; Gross et al. 2015; Fig. 10).

Conclusions

Organic geochemical analyses (TOC wt% and Rock–Eval pyrolysis) coupled with stable isotopic proxies were performed on the organic-rich shale and coal samples of the Early Paleocene (Danian) Hangu Formation in the

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Fig. 10 Block diagram illustrating depositional environment of the Hangu Formation (Shallow marine inner shelf setting)

Kala-Chitta Range of Northwest Pakistan to assess its source rock potential that resulted into the following conclusions;

- The Hangu Formation is lithologically heterogeneous and is primarily dominated by sandstone, limestone, laterite, organic-rich shale and coal lithofacies. The coal and organic-rich shale intervals are abundant in upper part of the formation and are assessed for hydrocarbon source rock potential evaluation.
- The organic matter encountered within Hangu Formation is mainly dominated by gas prone Type III kerogen with minor oil/gas prone Type II kerogens, which are thermally over-mature for liquid hydrocarbons but can generate gaseous hydrocarbons. The relatively lower HI and higher OI of organic matter demonstrated that the kerogens are largely dominated by terrestrial OM with minor marine organic influx. The organic geochemical proxies revealed that the Hangu Formation possesses poor to excellent source rock intervals in terms of organic-richness. The coal intervals hold good to excellent source rock quality, whereas the organic-rich shales acted as poor to moderate source rock.
- The organic geochemical data dechiphered an indigenous nature of the organic matter and hydrocarbons within the formation. In response to increased thermal maturation, the representative rock samples from the Hangu Formation have shown low potential to produce liquid hydrocabons. Thus, overall, the formation is thermally over-mature and can seldom generate liquid hydrocarbons but can act as a source for gaseous hydrocarbons. The over thermal maturity is most likely because of an effect attained from nearby tectonically active Himala-

yan Foreland Fold-and-Thrust Belt system and associated metamorphic sequences as a consequence of himalalyan orogeny.

• The depositional environment etablished for Hangu Formation exemplified a shallow marine proximal inner shelf setting with prevaliane of episodic sub-oxic to anoxic conditions that enhanced organic matter preservation and enrichment within the coal and organic-rich shale lithofacies as infered from organic gechemical and stable isotopic proxies.

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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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