

ORGANIC GEOCHEMICAL ASSESSMENT AND SHALE GAS POTENTIAL OF LOWER SILURIAN ORGANIC RICH SHALE IN THE GHADAMES BASIN, NORTH AFRICA

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Abstract. *The Silurian organic rich shale is the main source of hydrocarbons in the Ghadames Basin in North Africa. The basin has been widely characterized as a source rock for Ordovician oil and gas; yet understanding this shale as a shale resource play remains problematic and challenging. In this study, geochemical and mineralogical analyses of drill cuttings derived from five wellbores were carried out to evaluate the unconventional potential of Silurian organic rich shale.*

The results of geochemical analysis show that the present day total organic carbon (TOC) of this shale is generally medium to good, varying between 1 and 5 wt%. The hydrogen index (HI = 55–201 mg HC/g TOC) and Tmax (435–454 °C) values indicate type II kerogen in a mature state and its ability to generate wet gas. The results of mineralogical analysis show that clay minerals dominate in all samples of Silurian shale (39–58%) followed by quartz (16–37%). Geochemical parameters such as Mo, V, As, Zr and TiO₂ indicate that these shales were deposited in anoxic conditions and were sourced from intermediate igneous rocks. In order to evaluate the potential of Silurian shale as oil and gas source, in this work, the chemostratigraphy technique was applied to identify the provenance of silica in shales, to characterize the shale brittleness and model a correlation between the mineralogy and organic matter content. In conclusion, the Silurian organic rich shale in the Ghadames Basin exhibits good characteristics for shale resource hydrocarbons production.

Keywords: *Silurian organic rich shale, shale gas, chemostratigraphy, Rock-Eval pyrolysis, Ghadames Basin.*

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1. Introduction

Shale gas has become an important hydrocarbon target to substitute conventional oil and gas [1] owing to the high abundance of shale in the sedimentary basins worldwide [2]. For example, in North America the production of shale gas was more than 8 Bcfd (billion cubic feet per day) in 2009 [3], including the Marcellus, Haynesville, Eagle Ford and Barnett shale formations.

The Lower Silurian shales in the Ghadames Basin in North Africa have served as the most important petroleum source rock in the entire Saharan Platform [4]. The gross thickness of Silurian shale is more than 300 m; however, the thickness of organic-rich shale ranges from 10 to 65 m. The total organic carbon (TOC) of this type II kerogen marine shale averages 5% and the thermal maturity ranges from mature to postmature, placing the study area in the condensate and wet to dry gas window.

An earlier study of Lower Silurian organic-rich shale in the Ghadames Basin has estimated its gas and oil reserves at about 1020 Tcf and 110 billion barrels, respectively [5].

Thus, the successful development of shale gas resources can potentially enhance and expand the fossils energy exploration and exploitation in the region. The aim of this study is to characterize the geochemical composition of these Lower Silurian hot shales and to determine their mineralogical proprieties in order to define the shale's potential to produce shale gas. Furthermore, a comparison is made of our results with those obtained for the much known commercial gas shale systems in the USA.

2. Geological background

The study area lies in the Ghadames Basin, which is considered as one of the main oil and gas basins of North Africa. It covers large parts of Libya, Tunisia and Algeria on a total area of more than 350,000 km² [6]. The Lower Silurian shale is widely regarded as the main source rock in the Ghadames Basin [7]. This graptolitic radioactive Lower Silurian (Llandovery) black shale was deposited during a post-glacial marine transgression (Fig. 1). Locally, this radioactive shale is called "Tanezuft" [8, 9].

The Tanezuft-Ghadames Total Petroleum System is an important total petroleum system with regard to known volumes of oil and gas, containing about 30% of the discovered oil and 60% of the discovered gas in the province. The Lower Silurian Tanezuft shale has a mean TOC in the range of 0.5–2% (individual values may reach 17%) and originally contained type II kerogen which was predominantly derived from marine plankton [10–12]. The original hydrogen index (HI) is estimated to near 600 mg HC/g TOC [10]. However, in the present day, the Silurian shale in the study area is mostly late- or postmature, and consequently has lower HI [13].

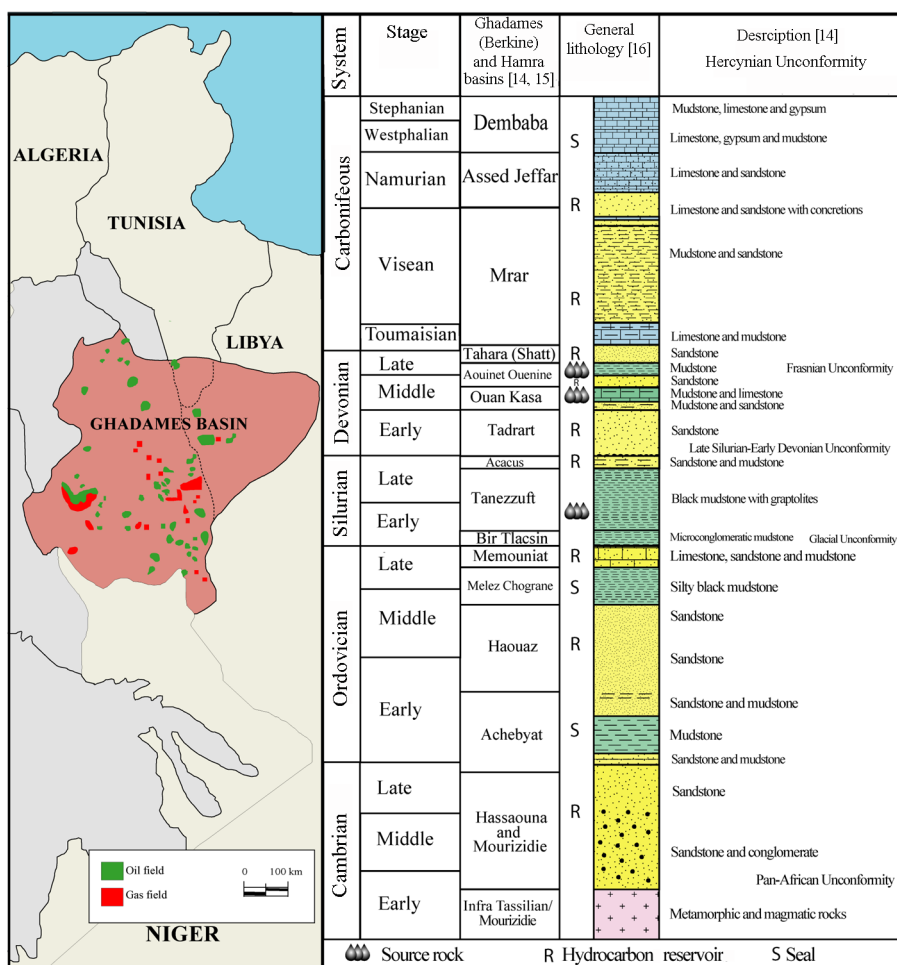


Fig. 1. Location map and stratigraphic nomenclature of the zone of interest within the Ghadames Basin (modified from [14–17]).

Tissot et al. [7] and Daniels and Emme [10] suggested that the oil generation from Silurian source rocks probably started and reached peak generative phases in the Carboniferous, but it was stopped during the Hercynian deformation phase (Fig. 2). At present, the Silurian source rocks are in the wet to dry gas generation phase [10].

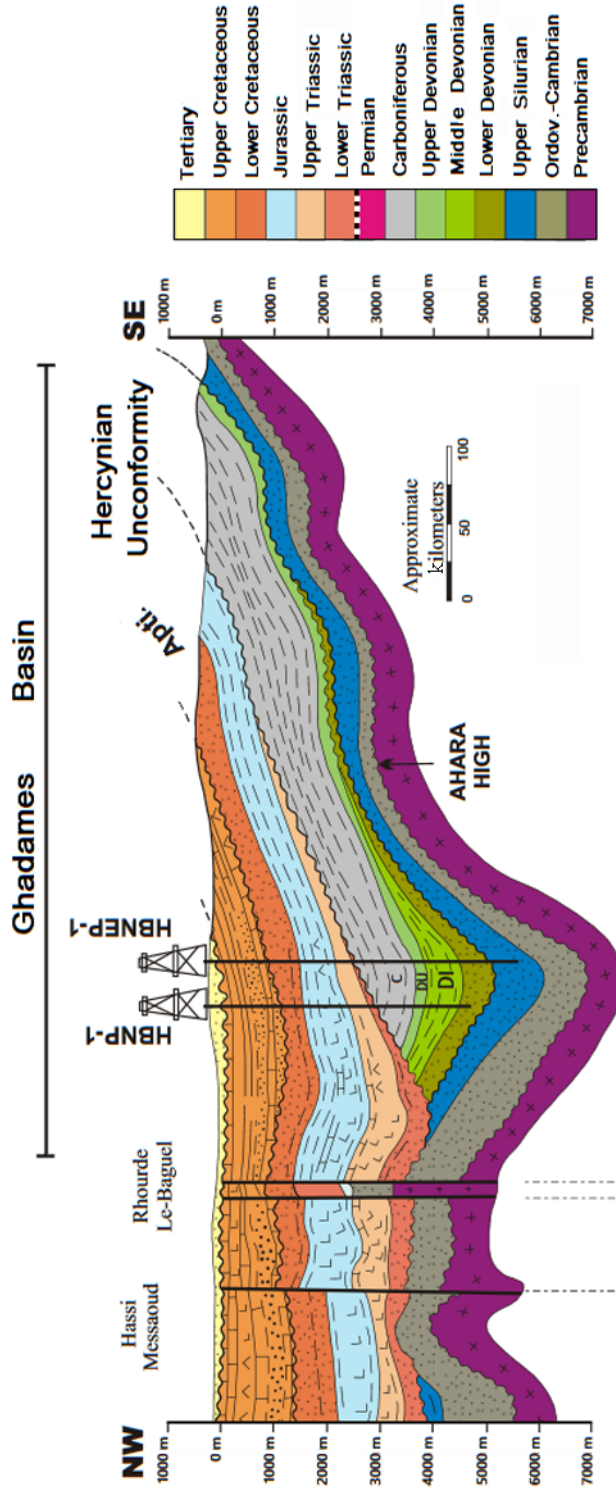


Fig. 2. Stratigraphic cross sections through the Ghadames Basin ([18], modified from [19]). (The abbreviations used: Apti. – Aptian; Ordov. – Ordovician).

3. Materials and methods

Overall 22 samples covering Tannezuft black shales were collected from five different wells in the Ghadames Basin. The samples were subjected to Rock-Eval pyrolysis, X-ray fluorescence (XRF) and X-ray diffraction (XRD) analyses in order to evaluate the organic matter content, maturity stage and mineral composition of Silurian organic-rich shale.

3.1. Geochemical analyses

The drill cuttings were collected from wellbores drilled with water-based mud to avoid any contamination effect of diesel and mud additives present in oil-based mud. Moreover, all non-organic carbons were removed by acidizing the samples with hydrochloric acid before performing the analyses. During pyrolysis, free hydrocarbons and hydrocarbons-like compounds were evaporated and quantitatively determined as S1 peak. However, the non-volatile organic matter cracked and the mobilized hydrocarbons and hydrocarbons-like compounds were measured as S2 peak. T_{\max} value corresponds to the temperature at which the maximum release of hydrocarbons from cracking of kerogen occurs during pyrolysis [20].

3.2. Mineral and elemental composition

XRF and XRD analyses were carried out on powder samples in order to determine the elemental and mineral composition of Silurian organic-rich shale. We used an Olympus XRF to provide the chemical composition and a BTX XRD to identify and quantify the different minerals in the samples based on their diffraction pattern. Each crystalline compound had a characteristic diffraction pattern, allowing its identification through the angular positions and relative intensities of the diffracted peaks. The minerals were identified by comparing the XRD diffractograms with diffraction patterns of individual phases provided by the International Centre for Diffraction Data (ICDD).

4. Results and interpretation

TOC contents of Silurian shale samples are given in Table 1. These contents vary between 0.96% for sample 19 (well X-5) and 5.22% for the Tannezuft Formation (well X-2). The samples from the Tannezuft Formation show the highest organic carbon values, from 3.52 to 5.22%. Cuttings with TOC values greater than 4% are considered as organic-rich samples with very good petroleum potential [21], which confirms that the Tannezuft Formation is favorable for production and preservation of organic matter.

The Rock-Eval pyrolysis parameter S2 values vary between 0.82 and 6.32 mg HC/g rock, while S1 values are ranging from 0.36 to 3.19 mg HC/g

rock. The hydrogen index (HI) values of the studied samples are generally low, varying between 54 and 201 mg HC/g TOC, while the average oxygen index (OI) is almost 9 mg CO₂/g TOC.

Table 1. Total organic carbon and Rock-Eval pyrolysis data for Silurian shale in the Ghadames Basin

Well No.	Depth,m	TOC, %	S1, mg HC/g rock	S2, mg HC/g rock	T _{max} , °C	HI, mg HC/g TOC	OI, mg CO ₂ /g TOC	S1/COT	PI
X-1	1800	1.15	0.45	0.99	442	86.09	9.00	39.13	0.31
X-1	1840	1.45	0.65	1.49	444	102.76	10.00	44.83	0.30
X-1	1900	2.14	1.19	2.15	445	100.70	9.29	55.74	0.36
X-1	1930	4.14	2.19	4.55	448	110.04	8.69	52.96	0.32
X-2	1760	1.52	0.68	1.04	434	68.42	9.46	44.74	0.40
X-2	1800	1.65	0.57	0.90	435	54.55	10.48	34.55	0.39
X-2	1875	2.31	0.68	1.44	439	62.47	10.13	29.50	0.32
X-2	1910	2.35	2.38	4.73	444	201.71	10.24	101.49	0.33
X-2	1950	5.22	3.19	6.32	445	121.19	8.45	61.17	0.34
X-3	1810	1.09	0.36	1.19	439	109.68	10.45	33.18	0.23
X-3	1850	1.39	0.49	1.38	439	99.64	10.14	35.38	0.26
X-3	1900	2.04	1.15	1.99	439	97.79	9.35	56.51	0.37
X-3	1920	3.57	1.39	2.80	440	78.43	8.78	38.94	0.33
X-4	1760	1.02	0.68	1.04	430	101.96	9.57	66.67	0.40
X-4	1800	1.06	0.57	0.90	435	85.31	8.56	54.03	0.39
X-4	1875	1.91	0.68	1.44	438	75.59	7.53	35.70	0.32
X-4	1930	2.05	1.38	1.73	436	84.60	8.05	67.48	0.44
X-5	1800	1.01	0.47	1.14	450	112.87	13.86	46.53	0.29
X-5	1850	0.96	0.43	0.82	447	85.42	8.33	44.79	0.34
X-5	1890	1.64	0.55	1.01	449	61.59	9.15	33.54	0.35
X-5	1900	1.79	0.62	1.22	453	68.16	4.47	34.64	0.34
X-5	1905	3.52	2.95	5.72	454	162.50	9.39	83.81	0.34

4.1. Organic matter richness and hydrocarbon generation potential

The S2 vs TOC diagram (Fig. 3) shows that the majority of the samples are rich in organic matter whereas the petroleum potential is considered low to medium, except for samples of the Tannezuft Formation whose values of S2 may reach 6 mg HC/g rock. So the petroleum potential of Tannezuft source rock is considered medium to good with TOC ranging from 2 to 6%.

The high S1 values suggest potential source rocks. However, high S1 values can also result from contamination with migrated oil or drilling fluids additives [21]. Hunt [22] asserts that nonindigenous hydrocarbons can be identified whenever S1 is high and TOC low.

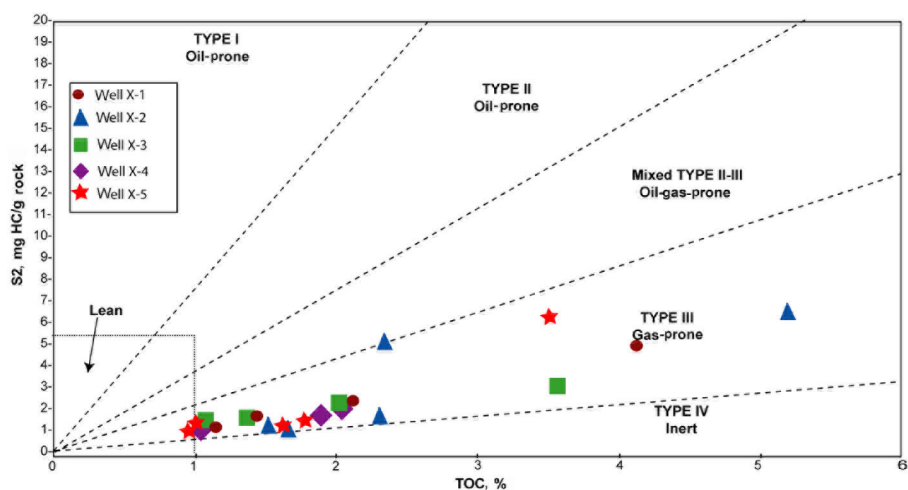


Fig. 3. S2 vs TOC diagram. Tannezuft shales are considered rich in organic matter with good oil potential.

We used the TOC vs S1 diagram to differentiate indigenous hydrocarbons from non-indigenous ones for the Silurian shale samples. Accordingly, all the samples indicate the presence of indigenous hydrocarbons and no external contribution of migrated hydrocarbons (Fig. 4).

Combining the data for S1 and S2 in the S1-S2 diagram allows the characterization of petroleum potential of the studied samples (Fig. 5). The majority of the samples have a medium to good petroleum potential, except those from the Tannezuft Formation, which are characterized by high petroleum potential. These results correlate with the HI values given in Table 1. All samples are in the production index (PI) range between 0.1 and

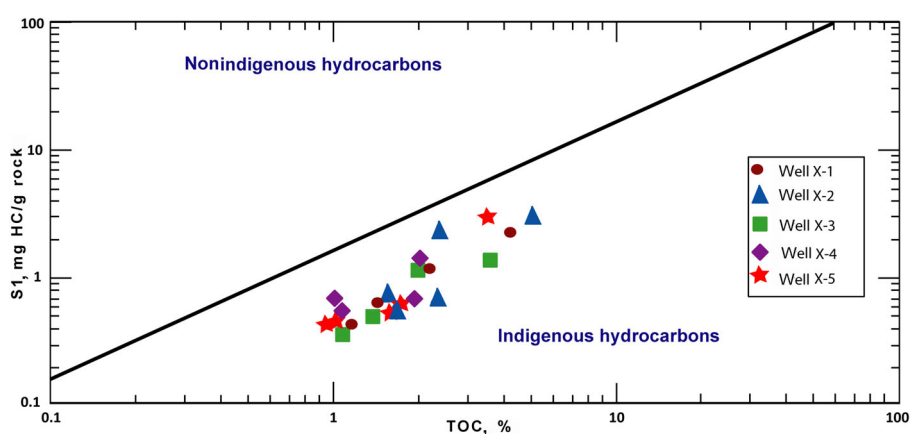


Fig. 4. TOC vs S1 diagram identifying migrated hydrocarbons in Silurian shale.

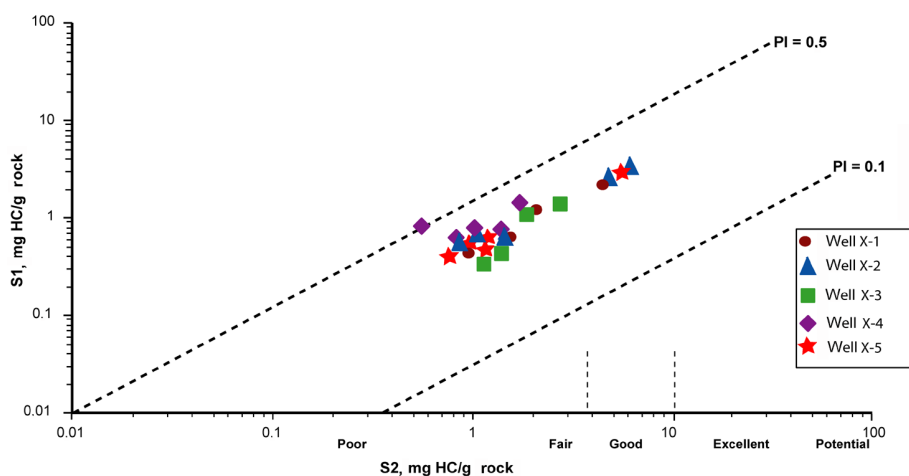


Fig. 5. Petroleum potential of Silurian shale in the Ghadames Basin.

0.5 indicating an oil and gas window. Typically, shale gas production beds show PI values of 0.5 to 1.5, while shale samples with a PI greater than 0.1 can generate an excellent amount of hydrocarbons.

4.2. Type of organic matter and hydrocarbon generation potential

The hydrogen index is defined as $[(100 \times S_2)/\text{TOC}]$. In the studied samples, the values of HI range from 54.5 to 201 mg HC/g TOC and those of T_{max} are between 435 and 454 °C (Table 1). The HI vs T_{max} diagram shows that the majority of the samples are in the oil window with a clear trend towards the gas window for samples from wells X-1 and X-5.

According to Hunt [22], a hydrogen index value greater than 200 mg HC/g TOC implies a high oil generation and expulsion potential. On the other hand, if the values are lower than 150 mg HC/g TOC, the gas is then the main fluid generated. Thus, based on Figure 6a, we can assert that in Silurian clays and especially the Tannezuft Formation, gas would be the main product generated, since almost all HI values are lower than 150 mg HC/g TOC.

The HI vs T_{max} plot (Fig. 6a) proposes two types of organic matter: terrestrial (Type III) and marine (Type II). The distribution of HI values between type II and III kerogen domains suggests that several samples contain varying proportions of these two types of organic matter. In addition, it is noted that the hydrogen index decreases as maturity increases.

The variations of HI vs OI (Fig. 6b) indicate that the majority of the Silurian shale samples are placed between the type II and type III kerogen lines with a tendency toward type II kerogen. This confirms the marine and planktonic origin of organic matter.

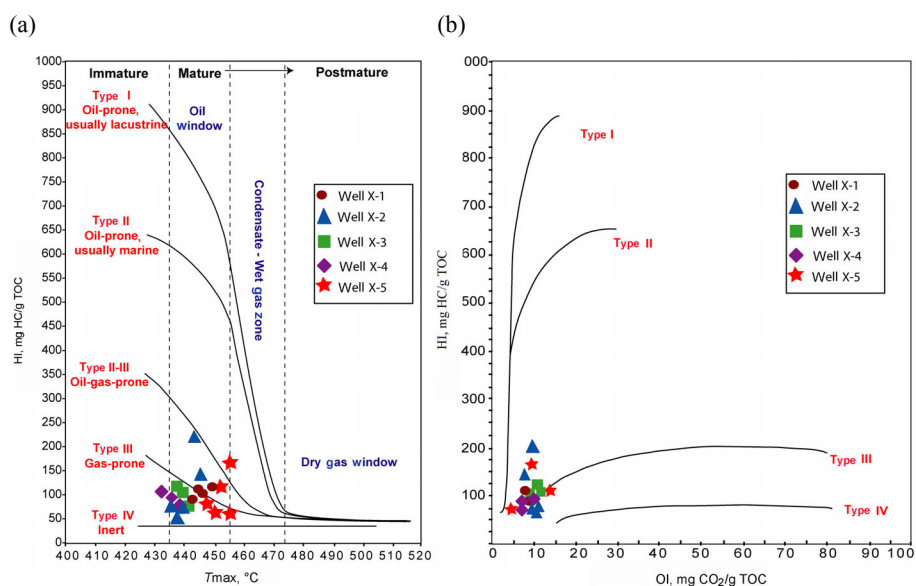


Fig. 6. a) HI vs T_{max} and b) HI vs OI cross-plots to show the kerogen type and maturity degree of Silurian shale samples.

4.3. Elemental composition and mineral content of Silurian shale

The chemical composition of the studied shale (Table 2) shows its silica to be of terrigenous origin, with no biogenic silica involvement (Fig. 7a) [23]. The chemical classification of siliciclastic sediments (Fig 7b) reveals that the majority of the studied samples belong to shales with few analyses indicating the wacke composition. The variation of Zr and Ti suggests that most Tannezuft shale samples have an intermediate provenance between the mafic and felsic sediments (Fig. 7c) [24].

Table 2. The results of total elemental analysis of studied shale samples, %

Well No.	Depth, m	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	K ₂ O	CaO	SO ₃	P ₂ O ₅	TiO ₂	MnO
X-1	1800	54.59	19.41	8.82	2.60	1.81	4.47	1.22	1.28	0.03
X-1	1900	45.78	18.76	11.89	2.54	1.94	9.61	0.68	0.80	0.02
X-2	1760	66.13	15.23	6.33	3.16	1.51	0.83	1.79	1.07	0.06
X-2	1800	52.71	21.35	10.22	2.77	0.82	5.79	1.21	1.13	0.04
X-2	1875	53.31	19.54	10.31	2.57	1.84	5.72	1.14	1.10	0.03
X-2	1910	51.34	19.76	9.48	3.15	2.93	5.32	1.12	0.94	0.03
X-2	1950	48.01	18.06	11.01	2.46	2.55	8.19	0.89	0.89	0.02
X-3	1810	55.41	18.23	8.54	2.56	2.03	4.24	1.21	1.24	0.03
X-3	1760	58.40	15.49	9.72	2.32	1.65	6.27	1.07	1.48	0.03
X-3	1850	50.12	18.52	9.96	2.44	2.69	5.33	1.08	1.08	0.03
X-3	1900	47.56	18.33	11.61	2.44	2.49	6.13	0.89	0.94	0.02
X-3	1920	45.78	18.76	11.89	2.54	1.94	9.61	0.68	0.80	0.02

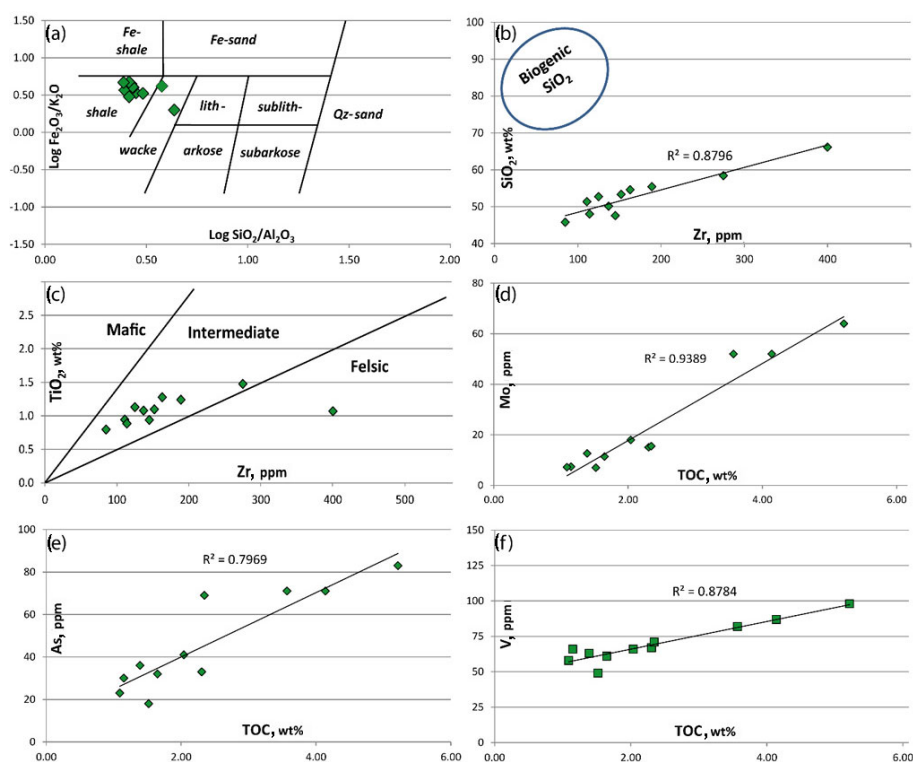


Fig. 7. Geochemical characteristics of studied shale samples: a) SiO_2 vs Zr plot characterizing silicon provenance; b) Herron's diagram identifying lithology and prevalence of shale; c) Hayashi diagram TiO_2 vs Zr identifying the provenance of sediments compounds; d) Mo vs TOC plot showing the positive correlation between organic matter and anoxic key elements, $R^2 = 0.9389$; e) As vs TOC plot showing the correlation between organic matter and anoxic key elements, $R^2 = 0.7969$; f) V vs TOC plot showing the correlation between organic matter and anoxic key elements, $R^2 = 0.8784$.

The elevated concentrations of molybdenum (Mo), vanadium (V) and arsenic (As) in these sediments suggest deposition under anoxic conditions. The positive correlation between the abovementioned elements and TOC (Fig. 7) has been studied in detail by Tribouvillard et al. [25, 26] and Negri et al. [27] and further confirms deposition under anoxic conditions.

The XRD analysis demonstrates that the Tannezuft shale has a complex mineralogy, including quartz, clay minerals, feldspars, calcite and Fe-oxides. These results can be used to identify the chemical and physical properties of the shale. For example, the presence of quartz facilitates its fracturing prior to production. Clay minerals dominate in all samples of Tannezuft shale, ranging from 39 to 58% (Table 3), followed by quartz whose content varies between 16 and 37%, while the contributions of feldspars, Fe-oxides and carbonate minerals are lower.

Table 3. Mineral composition of Tannezuft shale samples, %

Well No	Depth, m	Quartz	Clay	K-feldspar	Calcite	Fe-oxides
X-1	1800	23.20	52.40	14.00	0.70	9.70
X-1	1900	16.60	54.40	12.20	3.80	13.00
X-2	1760	37.25	39.45	16.30	–	7.00
X-2	1800	18.70	57.45	13.35	–	10.50
X-2	1875	21.64	53.21	12.80	1.30	11.05
X-2	1910	17.85	50.65	15.50	5.00	11.00
X-2	1950	19.28	51.27	12.60	5.00	11.85
X-3	1810	25.80	50.05	12.90	2.00	9.25
X-3	1760	31.64	45.34	11.42	0.80	10.80
X-3	1850	20.84	52.06	12.50	4.70	9.90
X-3	1900	18.60	52.40	12.30	5.00	11.70
X-3	1920	16.53	54.32	12.30	3.70	13.15

Note: “–” signifies no data.

4.4. Gas shale potential of Silurian shale in the Ghadames Basin

The geochemical analysis indicates that the Silurian organic-rich hot shale has a good gas generative potential. The thickness of the shale unit varies from 10 to 100 m, the richness in organic matter is good to excellent and TOC of this type II kerogen marine shale ranges typically from 2 to 6%. In regard to maturity, the Silurian shale is in a wet gas phase, so the gas and liquids could be produced. Our results correspond to the data reported earlier by Kaced and Arab [17], Belhaj Mohamed [28] and Troudi et al. [29].

The potential of a gas shale system is influenced by different factors, including [30]:

- organic richness, thickness and extent of shale;
- kerogen type;
- thermal maturity;
- mineralogy and rheology of shale significantly influencing fractability;
- existence of faults and fractures.

Quartz and carbonates are considered as brittle minerals which affect the shale fragility and have a positive influence on the development of hydraulic fractures prior to production. Based on the mineralogy of Barnett shale, Jarvie et al. [31] considered quartz to be the main brittle mineral which could reflect shale brittleness (BI_1). Wang and Gale [32] proposed to use quartz and dolomite as brittle minerals (BI_2) in some specific dolomite-rich regions:

$$BI_1 = \frac{\text{Quartz}}{\text{Quartz} + \text{Calcite} + \text{Clay}}, \quad (1)$$

$$BI_2 = \frac{\text{Quartz} + \text{Dolomite}}{\text{Quartz} + \text{Calcite} + \text{Clay} + \text{Dolomite} + \text{TOC}}, \quad (2)$$

where all components are expressed in wt%.

Based on X-ray diffraction data, we used these two expressions to calculate the brittleness index of Silurian shale. The brittle mineral content of the samples ranges from 17 to 37 wt% and the brittleness index BI varies between 0.22 and 0.5, with an average value of about 0.3 (Fig. 8a). According to the grading standard proposed by Goktan and Yilmaz [33], the brittleness of Silurian shale makes it amenable to fracturing. In fact, the presence of natural fractures is confirmed by several hydrothermal minerals and also by core analysis as shown in Figure 8a.

The characteristics of Silurian organic-rich shale in the Ghadames Basin, such as kerogen type, TOC content, thermal maturity, mineral composition and brittleness index, are similar to those of US Barnett shale. At the same time, in brittle minerals content and brittleness index, Silurian organic-rich shale is similar to Devonian Marcellus shale in USA (Fig. 8b).

5. Conclusions

Twenty-two Silurian shale samples from the Ghadames Basin were analyzed for their organic and mineral content to evaluate the shale gas potential of the formation.

The studied samples showed total organic carbon values between 1 and 6% indicating a good to excellent source rock. The hydrogen index-oxygen index diagram shows that kerogen type is mainly II, while the HI- T_{max} diagram classifies the kerogen types as a combination of types II and III. On the TOC vs S1 diagram, all the samples are in the indigenous hydrocarbons zone, suggesting that the source rock had not been affected by any external alteration.

The studied shale samples are shown to be in the wet gas window. The T_{max} values vary between 439 and 454 °C indicating an oil- and wet gas-prone stage.

Based on X-ray diffraction analysis, the mineral composition of Silurian organic-rich shale includes about 50% of clay minerals, followed by quartz whose content varies between 16 and 37%, while the contributions of feldspars, Fe-oxides and carbonates are lower.

The average brittleness index of the shale, based on mineral composition, is about 30%.

Geochemical ratios suggest that the Silurian shale might have been derived from intermediate source rocks and was deposited in an anoxic environment. All the previous analyses and interpretations indicate that the Silurian organic-rich shale is a potential target for shale gas.

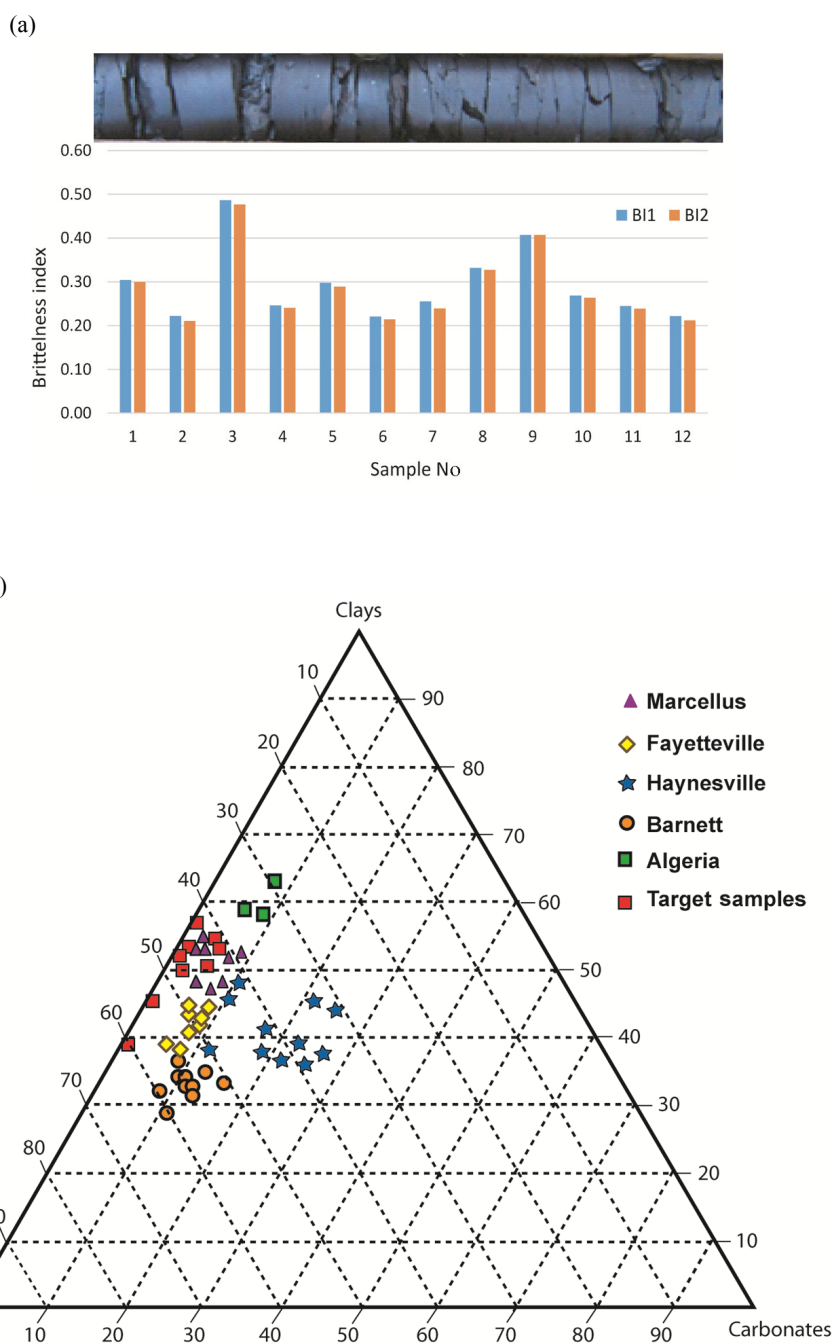


Fig. 8. a) Brittleness index of Silurian organic-rich shale and fractures in the shale core taken from well X-3 in the Ghadames Basin; b) comparison of mineralogies of Silurian Ghadames Basin shale and other shales worldwide.

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