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Organic Petrological and Geochemical Evaluation of Jurassic Source Rocks from North Iraq

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Abstract

Immature Jurassic oil shale is widely distributed and frequently outcropping in North Iraq. The organic-rich Jurassic sedimentary sequence, including prolific oil shale, was recorded in Banik area in Duhok Governorate of North Iraq. This sequence was systematically sampled from the geological formations; Sehkanyian, Sargelu and Naokelekan. The organic geochemical parameters were analyzed for 72 samples as well as one oil sample. A detailed study of petrologic properties was carried out for 12 samples. Based on TOC content, the Sargelu and Naokelekan formations can be considered as good to excellent source rocks, whereas Sehkanyian Formation has no potential since the TOC does not exceed 0.1 %. The samples of Sargelu and Naokelekan formations contain both kerogen types I and II indicating marine organic matter mainly derived from algae and phytoplankton organisms proposing typical oil prone source kerogen. This is further confirmed by the predominance of alginite and liptodetrinite macerals, where liptinite maceral group contribute more than 90% relative to other maceral contents. In general, Sargelu Formation samples have Production Index (PI), T_{max} and fluorescence parameters (λ_{max} and red/green quotient) suggesting immature to early mature stage of thermal maturity. The calculated ratios of Pr/Ph, Pr/nC_{17} and Ph/$nC_{18}$for the extracted bitumen and the oil sample, suggest generation of bitumen from marine organic matter deposited under reducing conditions at an early thermal maturity stage.

Keywords: Organic petrography, Geochemistry, Source rock, Jurassic, Kerogen, Bitumen, Iraq.

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1. Introduction

The Jurassic sedimentary sequence in northern Iraq contains very significant source rocks owing to their high total organic carbon (TOC) content, especially in both Sargelu and Naokelekan formations that were deposited throughout the Jurassic basin that exists in these areas [1-3]. As far as the authors are aware, the published data on these source rocks are insufficient and the available information concerning the TOC content, thermal maturation, and burial history require better understanding [4]. The dominant organic matter in oil shale is derived from one or more primary sources, such as terrestrial plants, lacustrine algae and marine organisms. These include large lake basins of tectonic origin, bogs, small lakes, lagoons and shallow seas [5].

Banik section is situated in the far north of the Iraqi territory, about 25 km to the NE of Zakho town, (Duhok Governorate). The studied section lies on latitude 37°13′ 33.4" N and longitude 42° 58′ 2.6" E, to the west of Banik village which is also known among resident farmers as Banik Haji Ghazi. Several mountains surround the area such as; Kokharash to the north, Khamtur to the west and Shaban to the south (Fig. 1). The present study deals with the hydrocarbon potential of the oil shale of the Sargelu and Naokelekan formations and their economic importance.

2. Stratigraphy

The Jurassic rocks are commonly exposed as isolated patches at some eroded cores and limbs of anticlines in the highly folded, imbricate and thrust zones of Northern Iraq. The sedimentary section in Banik area lies in a rugged terrain where the Jurassic rocks crop out in the southern limb of Kokharash anticline (mountain). The section is located close to the west of Banik village near the open coal mine (dug in rocks of Naokelekan Formation). The general trend of strata is E-W. The stratigraphic succession starts with Sarki and Sehkanyian formations (Early Jurassic) which underlies Sargelu Formation (Middle Jurassic). The latter is overlain by the younger Naokelekan and Barsarin formations (Late Jurassic), with Chia Gara Formation topping them (Fig. 1).

The lithologic composition of the Sargelu Formation consists of thinly bedded black bituminous limestone, dolomitic limestone and black papery shale intercalated by thin black streaks of chert in its upper part. In Banik section, the thickness of Sargelu Formation is about 30 meters of which the limestone rocks are mostly dolomitized. In the subsurface sections, at the foothill and Mesopotamian zone of the unstable shelf, the thickness is considerably higher and varies between 250 and 500 m Jassim and Buday [6].

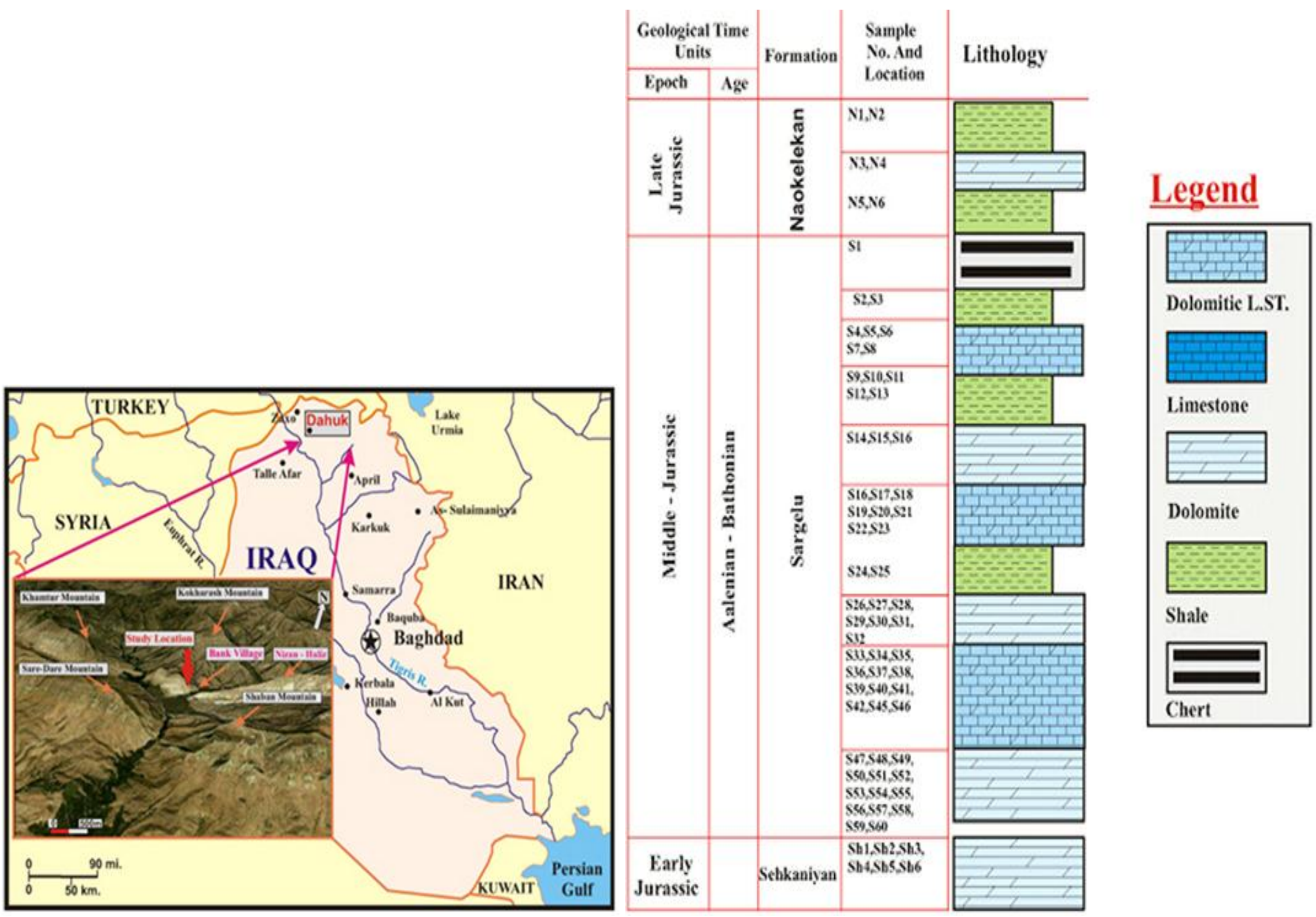

Fig-1. Location map and the stratigraphic column of Jurassic rocks crop-out in the Banik area, North Iraq indicating the collected samples.

According to Numan [7], the depositional history of the Sargelu Formation initiated and terminated as deep quiet marine environment, interrupted by some relatively shallow intervals but still remaining in the basinal realm. The organic–rich black shale and limestone of this Formation indicate anaerobic to dysaerobic conditions [8]. Such conditions are suitable for preservation of organic matter that gets thermally mature to generate hydrocarbon in subsequent geologic times. In some other subsurface sections, the behavior of the Sargelu Formation as potential source rock is provided by being seated at great depths [9].

The Naokelekan Formation consists of laminated bituminous limestone, alternated with bituminous shale (coal horizon) and thin bedded, highly bituminous dolomitie and limestone. Its age is assigned as Upper Oxfordian–Lower Kimmeridgian according to the study of ammonites of the Upper Jurassic [10]. The depositional environment of the formation was interpreted as euxinic in a slow subsiding basin [11]. The boundaries of the formation are agreed by many researchers to be conformable and gradational with underlying Sargelu Formation and overlying Barsarin Formation.

3. Methodology

Seventy two samples were collected to represent the targeted sedimentary interval in Banik area as well as one oil sample from Tawke oil field. The samples are collected along a traverse perpendicular to the bedding plane with spacing range of 0.5 to 1m. The collected samples represent the different lithologies of the Sargelu, Naokelekan and Sehkanyian formations. Analyses were conducted at Strato Chem Services© Company in Cairo, Egypt. The forty one samples that contain≥ 0.7% TOC, were analyzed by Rock-Eval pyrolysis. The measured parameters include; S1 (mg HC/g rock), S2 (mg HC/g rock), S3 (mg CO_2/g rock), T_{max} (°C), and TOC (wt. %) are quoted in Table 1. Several additional parameters such as Hydrogen Index (HI = S2/TOC*100), Oxygen Index (OI = S3/TOC*100) and Production Index (PI = S1/(S1+S2)) are calculated (Table 1).

Table-1. The ranges of the organic geochemical parameters of the Jurassic studied samples are summarized in the following table.

Formation	**TOC**	**S1**	**S2**	**S3**	**T_{max}**	**HI**	**OI**	**S1/ TOC**	**PI**
	Average	Average	Average	Average	Average	Average	Average	Average	Average
	(Min:Max)	(Min:Max)	(Min:Max)	(Min:Max)	(Min:Max)	(Min:Max)	(Min:Max)	(Min:Max)	(Min:Max)
U. Sargelu	4.12	0.21	37.17	1.53	443	366	39	4	0.02
	(0.06:19.52)	(0.03:1.05)	(0.8 : 127.16)	(0.08:4.89)	(437 : 449)	(96 : 697)	(3:165)	(1:16)	(0.00:0.06)
M.Sargelu	2.4	0.24	24.92	1.26	443	317	29	9	0.04
	(0.16:18.12)	(0.04:0.43)	(0.78:95.07)	(0.20:3.96)	(439:447)	(103:525)	(8:66)	(2:24)	(0.00:0.13)
L. Sargelu	8.82	0.72	62.62	1.83	443	446	32	6	0.02
	(0.12:28.57)	(0.04:2.45)	(1.49:192.89)	(0.45:3.78)	(441:446)	(57:690)	(6:92)	(2:12)	(0.00:0.03)
Naokelekan	17.77	3.34	98.94	2.2	441	524	15	18	0.03
	(5.39:25.55)	(0.78:6.42)	(18.05:149.1)	(1.41:3.70)	(434:445)	(335:635)	(8:32)	(13:25)	(0.02:0.04)
Sehkanyian	0.06								
	(0.03:0.09)								

Gas chromatography (GC) and Gas chromatography-mass spectrometry (GC-MS) were carried out to identify different hydrocarbon compound classes and biomarkers of the rock extracts and oil sample.

Organic petrologic examination of 12 samples was done, in the Kurdistan Institution for Strategically Studies and Scientific Researches, using DM 6000 Leica Microscope. Identification of the maceral types was carried out under reflected and blue light illumination. The latter is necessary to recognize the fluorescing liptinite macerals. The classification schemes proposed for the interpretation of organic materials by Teichmüller and Ottenjann [12], Teichmüller [13], Borchan and Powell [14], Scott [15] were adopted in the present study. The change in the intensities of the fluorescence spectra of liptinite group was measured during ultraviolet irradiation over a wavelength range from 400 to 700 nm.

4. Results and Discussions

Pyrolysis techniques were used to establish the hydrocarbon source and the likely hydrocarbon products or source type.

4.1. Organic Matter Richness

The TOC content of the study sediments can be considered as straight expression of kerogen and bitumen abundance. According to the classification given by Peters and Cassa [16], the upper part of the Sargelu Formation can be rated as having an excellent hydrocarbon potential (0.06% to 19.52%, averaging 4.12%) and the middle part as good hydrocarbon potential (0.16% to 18.12%, averaging 2.40%), whereas the lower part can be rated as an excellent hydrocarbon potential (0.12% to 28.57%, averaging 8.82%, table 1). The TOC contents of the Naokelekan Formation range from 5.39% to 25.55 %, averaging 17.77% and can be considered as an excellent source rock. The Sehkanyian Formation has no source potential.

4.2. Organic Matter Types

It is necessary to recognize, that the quantitative aspects of kerogen evolution vary from one type to the other as a result of differences in the original composition of kerogen and that these generalizations may not reflect the true nature of the chemistry that occurs during the maturation process [17]. So, the types of organic matter must be distinguished and identified because different types of organic matter have different hydrocarbon generation potential and products [5, 18, 19]. The data of Rock-Eval Pyrolysis (Tables 1) indicate that Sargelu Formation plots generally in the field of kerogen type I and II (Fig. 2) which suggest common lacustrine depositional environment. The organic matter is mostly derived from marine algae and phytoplankton organisms which can be considered as a typical "oil source" kerogen Dahl, et al. [20]. Only one sample lies in the field of kerogen Type III which is derived mainly from terrestrial organic matter. Naokelekan Formation belongs to kerogen type I and is considered as excellent oil prone. The upper part of Sargelu Formation is characterized by localized intervals with capacity for both oil and mixed oil/gas generation.

The oil-prone source rock intervals are characterized by very high TOC (5.37%-19.52%) and excellent potential to generate oil as indicated by their high Hydrogen Indices (pyrolysis S2 yields from 32 to 127mg HC/g Rock; and HI mostly >500 mg HC /g TOC). The mixed-prone source rock intervals are characterized by low to very high TOC (0.70%-8.13%) and fair to very good potential to generate oil and gas as indicated by their high Hydrogen Indices (pyrolysis S2 yields 2.60-19.03.16 mg HC/g Rock; and HI mostly >200 mg HC /g TOC).

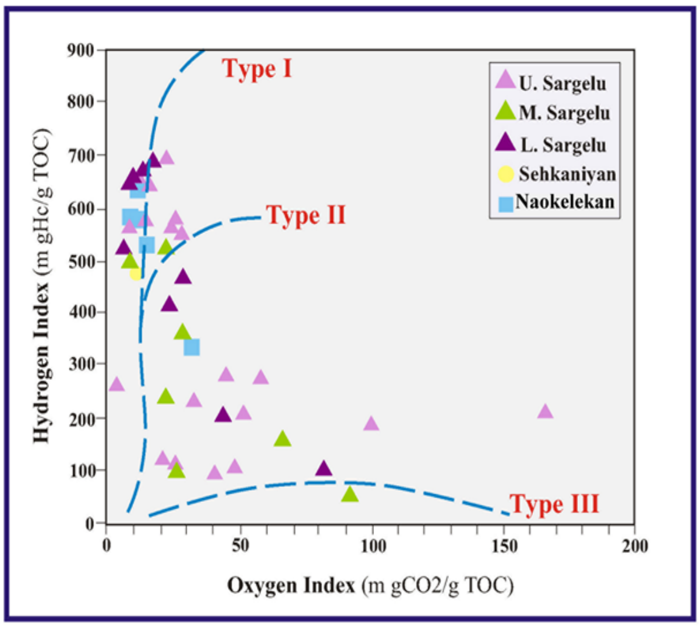

Fig-2. Van Krevelen diagram, showing Rock-Eval Hydrogen Index vs. Oxygen Index for the analyzed samples of the study area (adapted from Espitalié, et al. [21])

The middle part of the Sargelu Formation is characterized by having capacity for oil, mixed oil/gas and gas generation. The oil generating rocks are characterized by very high TOC and very good to excellent potential to generate oil as indicated by their high Hydrogen Indices (pyrolysis S2 yields from 17 to 95mg HC/g rock; HI 365-525 mg HC/g TOC). The mixed oil/gas generating rocks are characterized by high TOC (1.71%) and fair potential to generate oil and gas as indicated by their high Hydrogen Indices (pyrolysis S2 yields 4.15 mg HC/g rock; HI 242 mg HC/g TOC). In addition the gas generating rocks are characterized by relatively high TOC (1.81%) and fair potential to generate oil and gas as indicated by their high Hydrogen Indices (pyrolysis S2 yields 2.95 mg HC/g rock; HI 163 mg HC /g TOC).

Lower part of Sargelu Formation is characterized by having capacity for oil, mixed oil/gas and gas generation. The oil-prone source rocks are characterized by their very high TOC (2.34-28.57%) and very good to excellent potential for oil generation as indicated by their high Hydrogen Indices (pyrolysis S2 yields 14.75-192.89 mg HC/g rock; HI >400 mg HC /g TOC). The mixed oil/gas prone source rocks are characterized by very high TOC (4.29%) and good potential to generate oil and gas as indicated by their high Hydrogen Indices (pyrolysis S2 yields 8.83 mg HC/g rock; HI 206 mg HC /g TOC). In addition the gas-prone source rocks are characterized by very high TOC (3.28%) and fair potential to generate oil and gas as indicated by their high HI (pyrolysis S2 yields 3.50 mg HC/g Rock; HI 107 mg HC /g TOC).

Naokelekan Formation is characterized by having capacity for both oil and gas generation. The oil-prone source rocks are characterized by very high TOC (5.39-25.55%) and very good to excellent potential to generate oil as indicated by their high Hydrogen Indices (pyrolysis S2 yields 18.05-149.14 mg HC/g rock; HI >500 mg HC /g TOC, Fig. 3).

The plotting of S1 versus TOC can differentiate between the migrated and non-migrated hydrocarbons. The dividing line on the plot is where S1/TOC = 1.5. Values belonging to non-indigenous hydrocarbons plot above this line while indigenous hydrocarbon values emerge below it Hunt [22]. Thus all the samples analyzed of Sargelu and Naokelekan formations indicate indigenous hydrocarbons (Fig. 4).

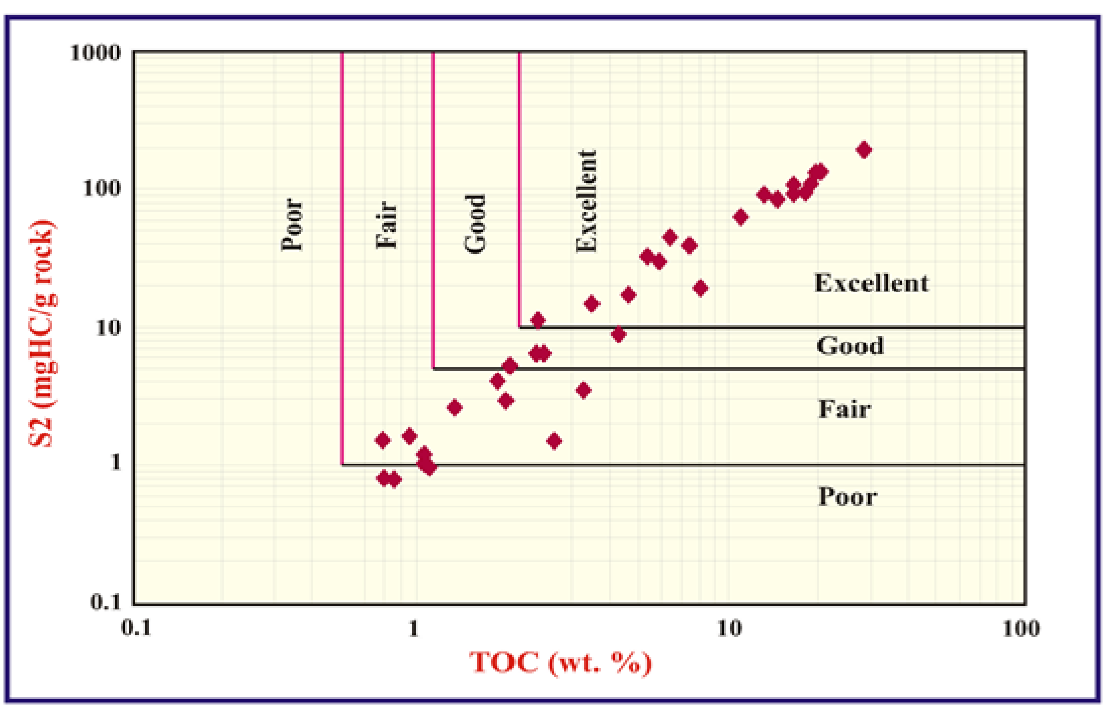

Fig-3. TOC wt% versus S2 plot of Sargelu and Naokelekan formations indicates their hydrocarbon potentialities.

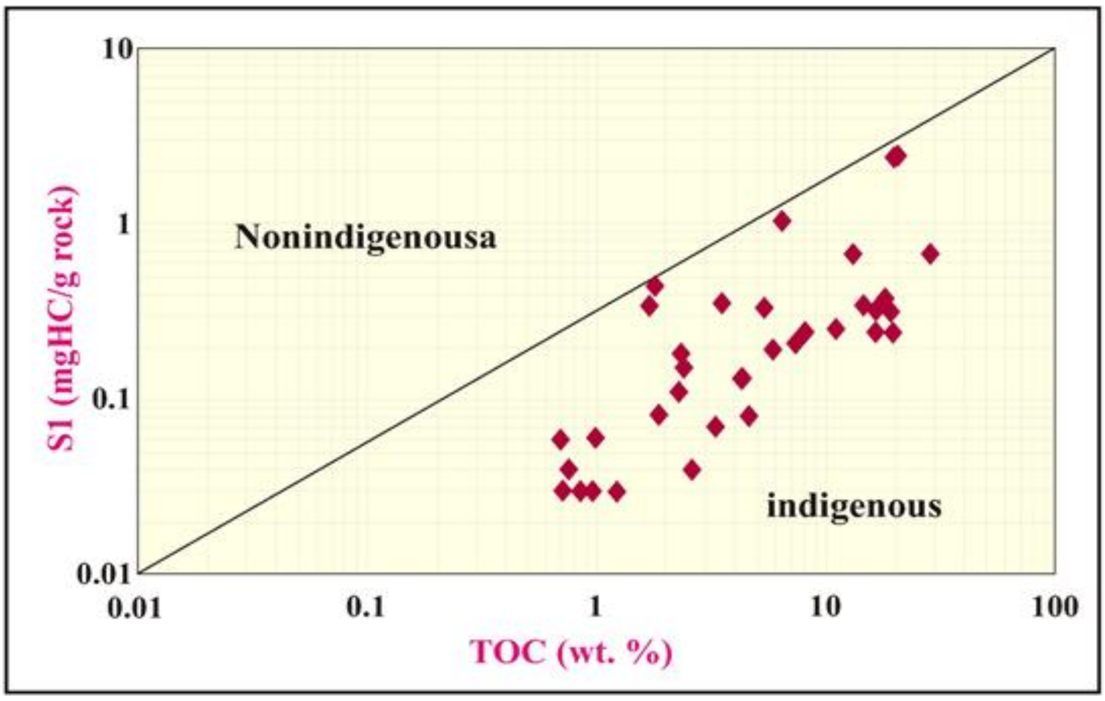

Fig-4. SI versus TOC for Sargelu and Naokelekan formations in Banik section (modified after Hunt [22])

4.3. Thermal Maturity

The maturity levels for the oil window depend on the type of organic matter [23], and encompass a vitrinite reflectance (Ro) ranges from 0.5 to 1.3% and temperature at maximum rate of hydrocarbon generation during S2 evolution (T_{max}) from 435 to 470°C. The PI parameter is another measure of maturity, with values ranging from 0.15 to 0.4 normally associated with oil generation. HI versus T_{max} is commonly used to avoid influence of the OI for determining kerogen type [22]. Determining kerogen type using HI versus T_{max} appears to be more accurate than OI versus HI. The difference between the results is expected because the Sargelu and Naokelekan formations are dominated by carbonates which affect the OI. The cross plot of HI versus T_{max} indicates dominance of kerogen typesI, II and mixed type II – III in both Sargelu and Naokelekan formations (Fig. 5). The samples are thermally immature to early stage of maturation, where pyrolysis T_{max} ranges from 437 - 449°C and PI is less than 0.15 [24]).

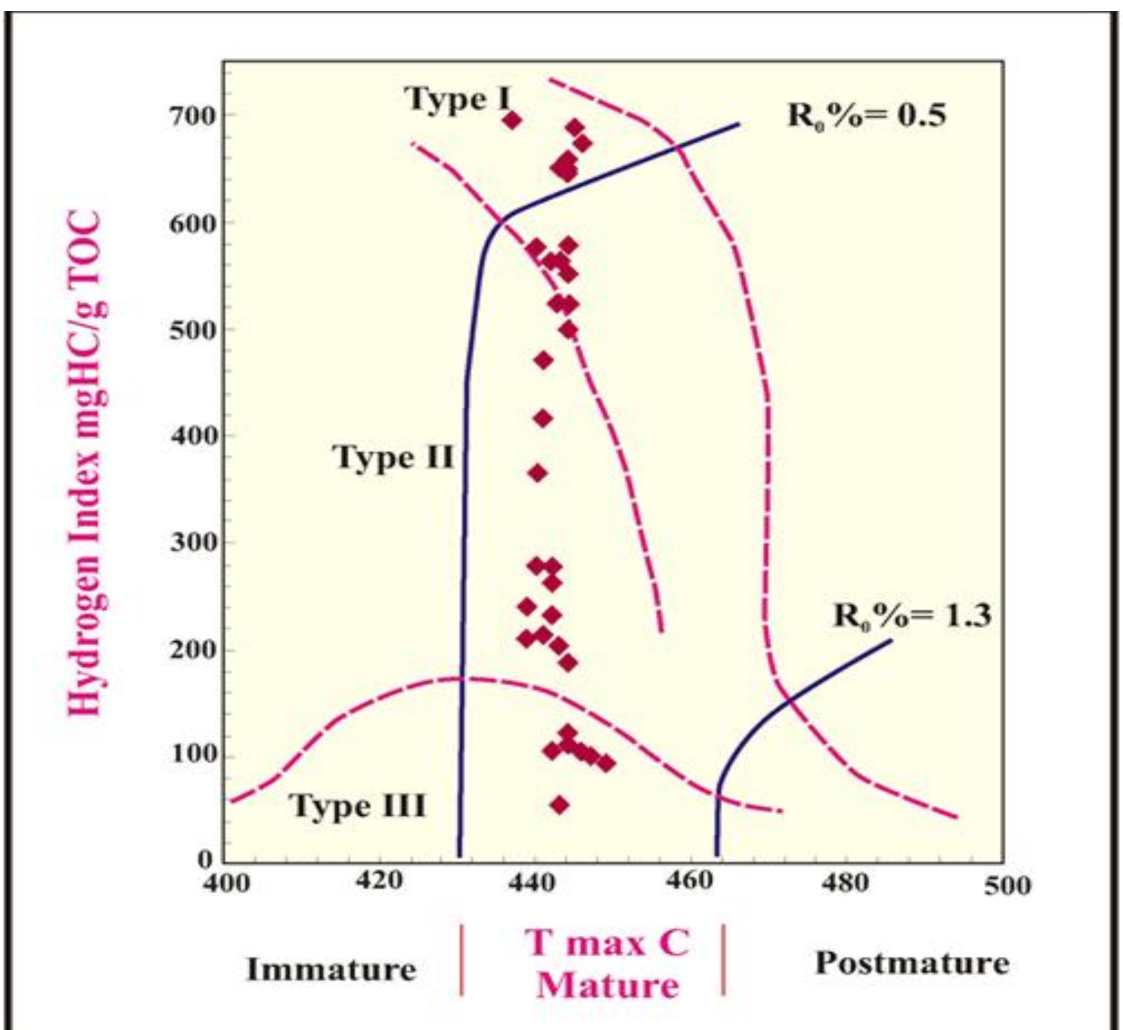

Fig-5. Kerogon plots for HI vs. T_{max} of the samples of Sargelu and Naokelekan formations of the Banik section.

4.4. Rock Extracts and Oil

Gas-chromatography analysis was performed for the extract of 6 rock samples in addition to one oil sample (Table 2).

Table-2. Gas Chromatograph data of the 6 extract samples of Sargelu Formation as well as the oil sample

S. No.	Pri/Phy	Pri/nC_{17}	Phy/nC_{18}	CPI Marzi[4]	N Paraffin	Isoprenoids	Resolved unknowns
Ext. 1	1.14	0.26	0.37	0.89	11.9	1.5	86.6
Ext. 12	1.33	0.39	0.39	0.91	12.7	2.0	85.3
Ext. 18	1.15	0.28	0.25	0.97	23.6	2.2	74.2
Ext. 21	0.91	0.24	0.29	0.87	25.3	2.9	71.7
Ext. 26	1.38	0.23	0.20	0.78	20.9	3.1	76.0
Ext. 52	0.70	0.28	0.44	0.95	23.5	3.6	72.9
oil	0.73	0.26	0.44	0.98	30.8	3.8	56.5

The gas chromatograms of the extracts show a rather unimodal distribution pattern of the normal alkane with maximum intensity light ones (<n-C20) indicating autochthones source of the organic matter (Fig. 6). The low content of hydrocarbon and the hump of the heavy hydrocarbon indicate that the samples are thermally immature. This is also certified by the low ratio of isoprenoid to normal alkane and the low PI parameter (< 0.10). The ratios of the Pristane/Phytane (Pr/Ph) in the 6 extracts are low (0.70 to 1.38), indicating anoxic reduced marine carbonate depositional environment. The organic-rich anoxic carbonate rocks generally generate oil with Pr/Ph ratio less than 2 [22, 25-27]. The carbon preference index (CPI) may be used as an indicator of maturation, where immature rocks often had CPI values more than 1.2 or less than 0.8. Some marine sponges, fresh water aquatic plants, ferns, fungi, yeasts and bacteria have small odd carbon preferences [28]. The average of the CPI of the extracts of Sargelu Formation is less than 0.9 indicating marine source at early stage of maturation.

The Pr/nC17 (0.23-0.39) and Ph/nC18 (0.20-0.44) ratios reflect deposition under reducing marine condition rather than effect of maturity or biodegradation (Fig. 7). The gas-chromatograms of the oil sample recovered from North Iraq at 2840 m of Sargelu Formation reveal n-alkane distribution pattern in the n-C_4 to n-C_{41} range (Fig. 6) with API value of 29.85º. The oil sample has low Pr/Ph ratio (0.73), which together with the Pr/n-C_{17} (0.26) and Ph/n-C_{18} (0.44) suggests generation from a source rock containing mainly marine organic matter deposited under reducing depositional conditions (Fig. 7).

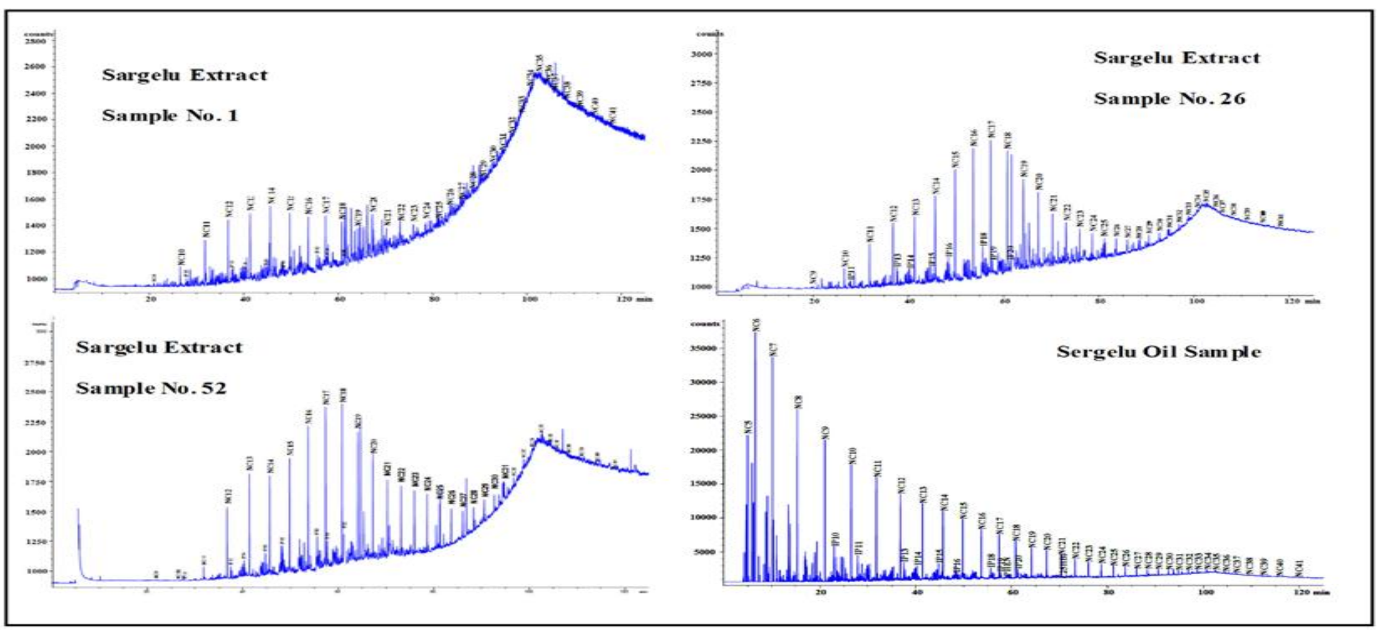

Fig-6. Gas Chromatograms of the rock extract of the Sargelu Formation and it oil sample from North Iraq.

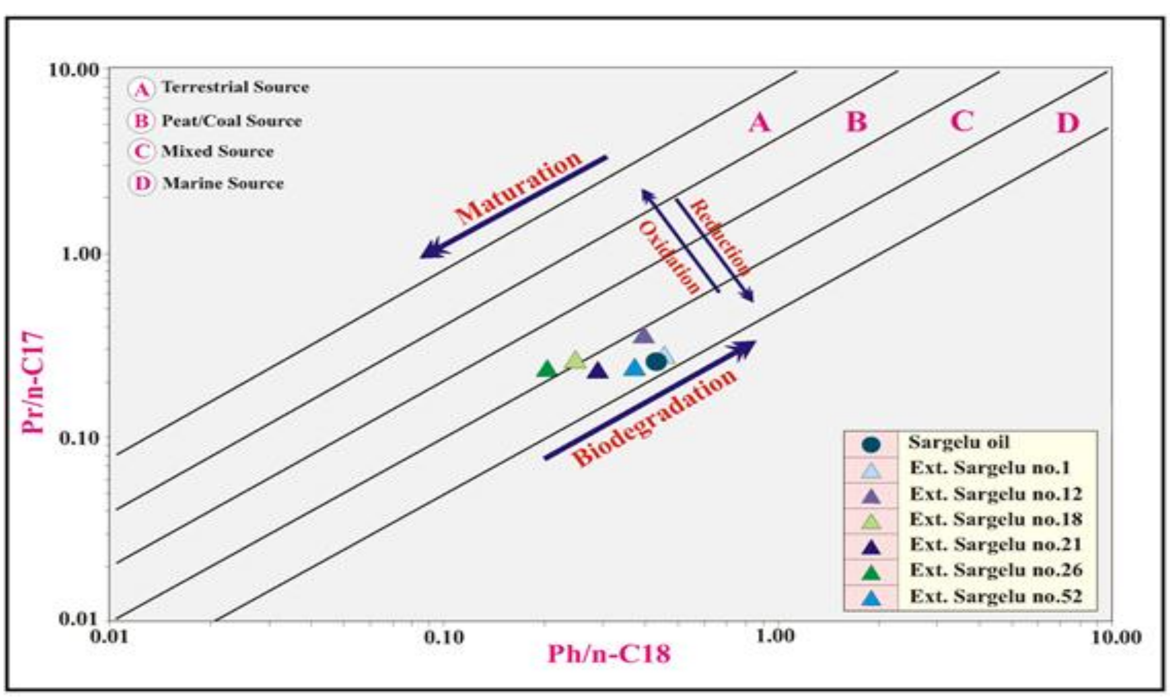

Fig-7. Pr/n-C17 versus Ph/n-C18 of the Sargelu Formation extracts and the oil samples.

4.5. Maceral Analysis

The majority of kerogen in the investigated samples belongs to liptinite maceral group (mostly of alginite and liptodetrinite), where the abundance of this group ranges from 85% to 99% relative to other maceral contents, on mineral matter free basis (Table 3).

Table-3. The maceral composition and the fluorescence properties of the oil shales of Sargelu Formation in Banik area, North Iraq.

Formation	Liptinite			Mineral	FluoresceneProperties	
	Alginite	Liptodetrinite	Bituminite	matrix vol%	λ_{max} (nm)	Q
	Average	Average	Average	Average	Average	Average
	(Min:Max)	(Min:Max)	(Min:Max)	(Min:Max)	(Min:Max)	(Min:Max)
U. Sargelu	45.67	21.67	32.67	70.00	520.00	0.66
	(45:46)	(20:24)	(30:35)	(68:74)	(515:525)	(0.60:0.72)
M.Sargelu	31	37	32	65	520	0.73
	(23:36)	(23:68)	(19:50)	(55:83)	(515:530)	(0.63:0.81)
L. Sargelu	54.33	15.00	30.67	57.33	526.67	0.63
	(45:60)	(13:17)	(25:40)	(56:58)	(520:530)	(0.62:0.64)

Blue light is necessary to recognize the fluorescing liptinite maceral as the major portion of the hydrogen-rich organic material, where it is invisible in normal reflected light (Fig. 8). It occurs in the form of thin wall layer with length ranging from 100 to 300 µm in the form of multifolded around foraminiferal chamber that is filled with framboidal pyrite and spheromorph bodies within the mineral matrix. The liptodetrinite macerals reach 10 µm in length and display fluorescence intensity similar to that of alginite (Fig. 8).

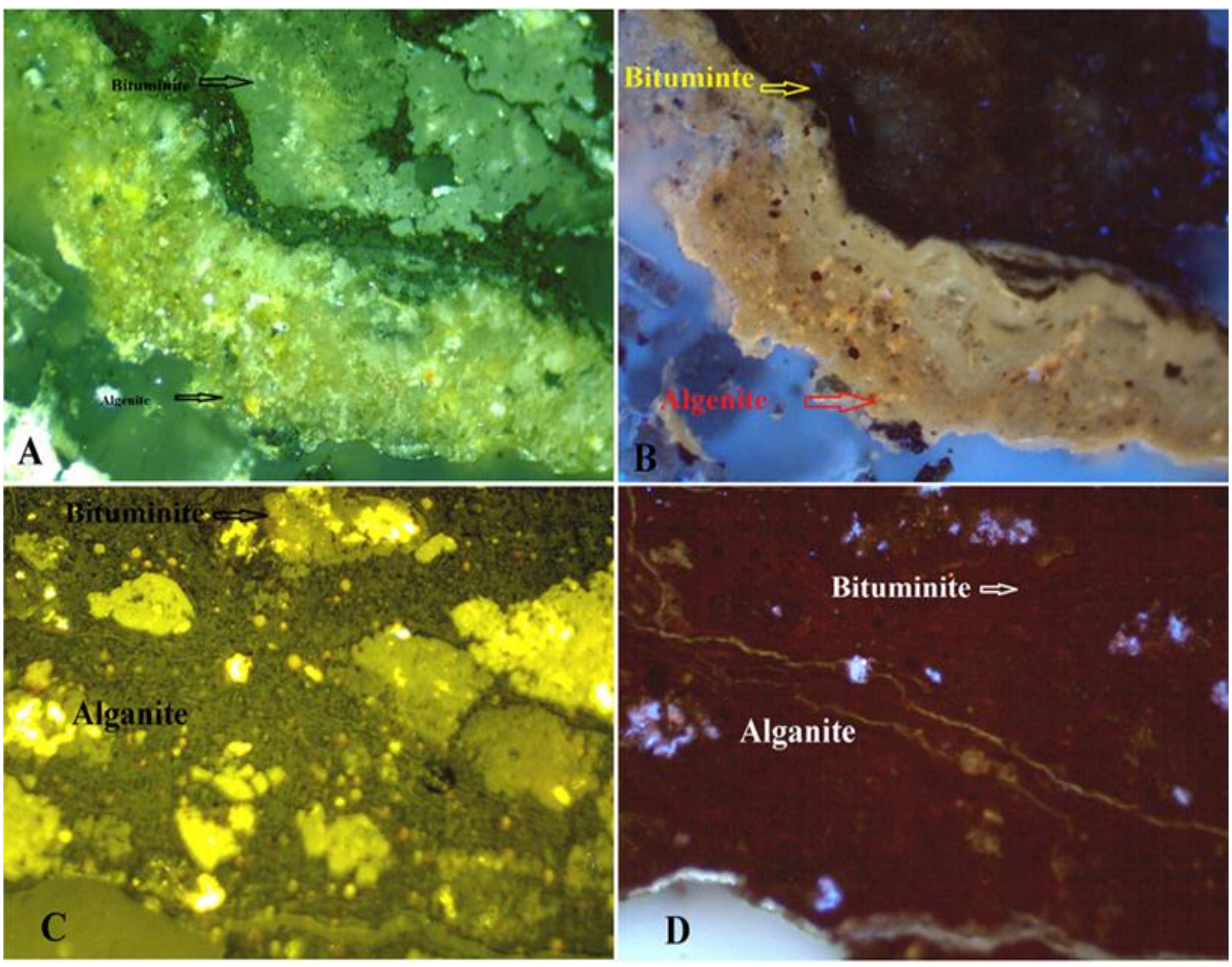

Fig-8. P Petrographic characteristics of Sargelu oil shale samples from Banik area, Northern Iraq. Alginites and bituminite A & C (Blue light) X200 B & D (Reflected light) X200

They were observed frequently in all samples. Liptodetrinite macerals cannot be assigned to a specific source because it consists of fragments of degraded remains of spores, cuticles, resinous bodies or algae [13]. The predominance of the liptinite maceral group and the presence of pyrite in all analyzed samples indicate organic material of marine origin deposited under anoxic environment. Bitumen like materials was frequently observed in all samples. According to Teichmüller and Ottenjann [12] bituminite macerals, which are decomposition product of algae, animal plankton and bacterial lipids, can be classified, in the present work, into two types; I and II. Bituminite "type I" occurs as lenticular bodies ranging from 100 to 150 µm in length and displays orange to brownish fluorescence in blue light. Bituminite "type II" displays petrographic characteristics similar to those of vitrinite. It occurs as oval or elliptical bodies with dark brown fluorescence color of very low intensity (Fig. 8). Quantitative methods have been developed to relate the changes in florescence properties (λ_{max} and Q values) to the geothermal rank [29]. Alginites are the best for rank evaluation, where cutinites and resinites are not suited for maturation evolution because they show varying fluorescence at a given rank stage [13]. The obtained data of λ_{max} values for alginite of the studied samples range from 515 to 530 nm and red/green quotient values range from 0.6 to 0.8. These values indicate low maturity level of the studied source rocks.

5. Conclusions

The Jurassic source rocks in North Iraq have been assessed for their hydrocarbon potential. A total of 72 rock samples from Sehkanyian, Sargelu and Naokelekan formations were analyzed by Rock-Eval /TOC pyrolysis and selected 12 samples were prepared for organic petrologic examination as well as 6 rock extract and an oil sample were analyzed by GC/GCMS.

In general, the Sargelu and Naokelekan formations have kerogen types I and II of lacustrine depositional environment in immature to early mature stage of thermal maturity. They can be considered as good to excellent source while the Sehkanyian Formation has no potential. The predominant macerals of the Sargelu Formation belong to liptinite group (alginite and liptodetrinite).The λ_{max} values for alginite of the studied samples and red green quotient values indicate low maturity level of the studied rocks.

The extracted bitumen samples from Sargelu Formation show diagnostic ratios of Pr/Ph, Pr/nC_{17} and Ph/nC_{18} for generation oil at an early thermal maturity stage. The low Pr/Ph ratio of the analyzed oil sample together with the low $Pr/n\text{-}C_{17}$ and $Ph/n\text{-}C_{18}$ suggest that the oil was generated from a source rock containing mainly marine organic matter deposited under reducing conditions.

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