Iranian Journal of Oil & Gas Science and Technology, Vol. 11 (2022), No. 1, pp. 1-15
http://ijogst.put.ac.ir

Investigation of Origin, Sedimentary Environment and Preservation of Organic matter: A Case Study in Garau Formation

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Highlights

- Quality and quantity of the organic matter in the Garau Formation according to pyrolyze and elemental analysis.
- The organic facies assessment using geochemistry parameters.
- The visual kerogen analysis and palynology

Received: June 22, 2021; revised: August 06, 2021; accepted: August 23, 2021

Abstract

Knowing the characteristics of suitable environments for precipitation of oil prone source rocks facilitates oil explorations and leads to development of oil fields. The current study investigates the organic matter properties and sedimentary environment conditions of the Garau Formation in various outcrop sections in Lurestan province from south-west of Iran (High Zagros) using elemental analysis, visual kerogen analysis and Rock-Eval pyrolysis data. The geochemistry parameters indicate the Garau Formation is an excellent oil prone source rock and composed of kerogen type I and II. The oxygen index (OI) is very low which reveals that organic matter deposited in an anoxic sedimentary environment and suitable for the preservation of organic matter and hydrocarbon generation. The visual analysis of isolated kerogens from source rock samples indicates the abundance of dark amorphous organic matter (AOM) with low amounts of phytoclasts and pyrite and not any palynomorph. Sedimentation seems to have occurred in deep and reduced parts of a carbonate basin during a rapid transgression. In addition, due to the effect of thermal maturation, the color of amorphous organic matter has darkened. The elemental analysis and Van-Krevelen diagram applied to show the type of organic matter and reveals the thermal maturity is related to the oil window. Elemental analysis reveals the high content of organic sulfur in the structure of kerogen. Moreover, the amount of pyritic sulfur (Sp) and organic sulfur (So) contents have been calculated.

Keywords: Elemental analysis, Garau Formation, Sedimentary environment, Visual kerogen analysis

How to cite this article
Hemmati .M, Ahmadi.Y, Investigation of Origin, Sedimentary Environment and Preservation of Organic Matter: A Case Study in Garau Formation, Iran J. Oil Gas Sci. Technol., Vol. 11, No. 1, p.1-15, 2022.
DOI: http://dx.doi.org/10.22050/ijogst.2021.291679.1601,

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1. Introduction

The increasing need for fossil fuel resources has led to petroleum geologists to perform various analyzes for oil explorations. Oil exploration programs are often complex and costly. However, determining the appropriate geological locations reduces the risks and costs of exploration (England, 2007). One of the basic conditions for formation and accumulation of hydrocarbons in reservoirs is the presence of a source rock with sufficient width and thickness, and thermal maturity (Behar et al., 2001). The important sedimentary environments for precipitation of potential source rocks are anoxic basins, which could be traced by Rock-Eval pyrolysis, elemental analysis and visual kerogen analysis. The elemental analysis by elemental separations and determination of their percentages has useful applications in detecting the kerogen type and organic matter maturity (Hunt, 1996; Van Krevelen, 1984; Bernard et al., 2010; Peters, 1986; Peters et al., 2012). Palynology of isolated kerogens on the thin sections applied to study of faunal changes, ratio and types of different organic matter with semi-quantitative methods (Suarez-Ruiz, 2012). Using the elemental analysis along with the Van-Krevelen diagram it's possible to examine the level of thermal maturation and sedimentary environment conditions (Hunt, 1996; Van Krevelen, 1984; Staplin, 1969; Thompson and Dembicki, 1986). The quality of organic matter in the source rock can be determined based on hydrogen index (HI; mg HC/g TOC) and oxygen index (OI; mg CO2/g TOC) or according to H/C versus O/C ratio in the Van-Krevelen diagram (Hunt, 1996; Van Krevelen, 1984). As well as, the quantity of organic matter evaluated based on the TOC (total organic carbon) by the Leco method (Bernard et al., 2010; Peters, 1986).

The Upper Jurassic-Tertiary sedimentary succession of the South Dezful Embayment is the host for most of the oil and gas reserves in the southwest of Iran. The Garau Formation Known as the Early Cretaceous Petroleum system, in which excellent source rocks were deposited in the lower part of the Garau Formation, during the BerriasianValanginian period (Bordenave and Burwood, 1990; Bordenave and Hegre, 2010). Due to the importance of the Garau Formation in petroleum geology of Iran, it has always been the case of petroleum geology and palynology studies. So that, reports state the Garau Formation is a possible source rock which charged limited reservoirs, and has composed of black organic matter rich facies with high hydrocarbon potential (Bernard et al., 2010; Bordenave and Burwood, 1990; Bordenave and Hegre, 2010; Ezampanah et al., 2013; Mahbobipour et al., 2016; Sarfi et al., 2015). The purpose of this paper is to investigate the origin and preservation of organic matter and to investigate the organic facies and sedimentary environment conditions of the Garau Formation in the Lurestan province, Alligudarz region, in south-west of Iran (High Zagros).

2.Geological setting

The study area is located in the Lurestan province, Alligudarz region, from south-west of Iran, High Zagros. The Zagros fold-thrust belt is the deformed state of the Zagros sedimentary basin (Mahbobipour et al., 2016), and limited to the Zagros reverse fault in northeast and to Dezful Embayment in southeast (Figure 1) (Bordenave and Burwood, 1990). The Lurestan basin is the northwest part of Zagros orogeny Belt (Alavi, 2007; Farzipour-Saein, 2009). After deposition of the thick Gotnia Anhydrite during the Late Jurassic, the deposition of the low energy Garau facies from Valanginian to Aptian times and even up to the Coniacian in the axial part of the Lurestan depression was occurred (Figure 2) (Wynd, 1965). The type section of the Garau Formation was measured on the southwestern flank of Kabir Kuh in southwestern Lurestan about 10 km northeast of the village Qaleh Darreh (Motiei, 1993). At type section the dark brown radioactive laminated pyritic marls alternate with fine-grained argillaceous limestones (Bordenave and Burwood, 1990). The formation overlies disconformably the Gotnia anhydrite and underlies the Sarvak Formation in its type section but it is different in various parts of Iran (Wynd, 1965). Figure 1 shows the study area and related tectonic elements.

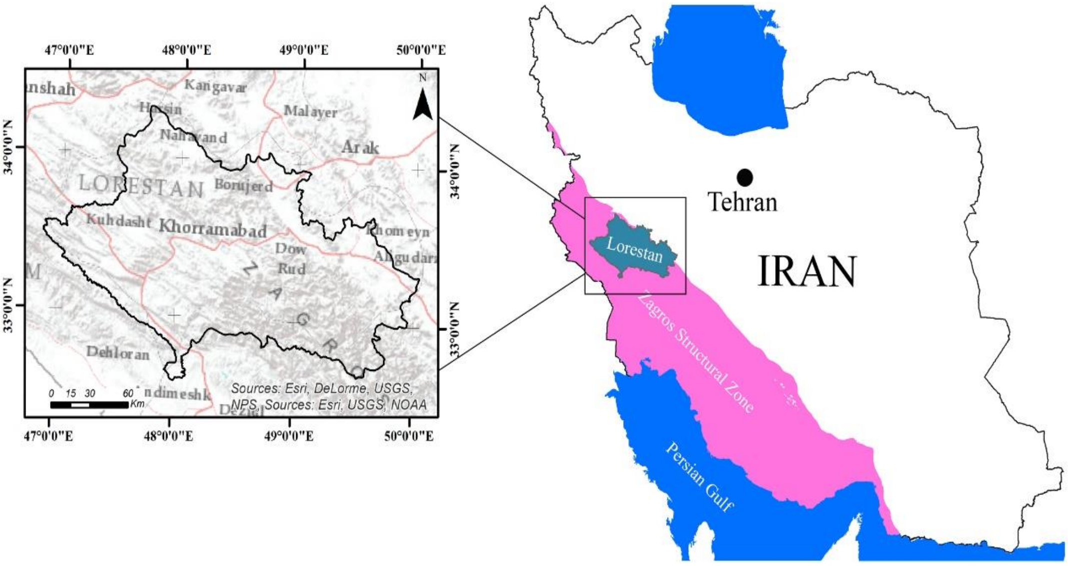

Figure 1
Structural setting of the Zagros Fold-Thrust-Belt showing the study area position.

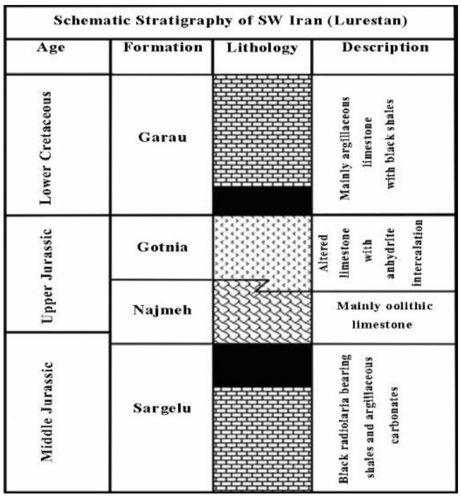

Figure 2
The schematic stratigraphy of Lurestan basin (Motiei, 1993).

3. Materials and methodology

In the current study, 123 surface rock samples have been collected from five outcrops (A, B, C, D, and E) of argillaceous limestones of the Garau Formation to obtain the pyrolysis data with Rock-Eval VI instrument for determining the total organic carbon (TOC) contents by the Leco et al method. In order to obtain the constituent elements by their percentages in the organic matter, the elemental analysis has performed on three isolated kerogens from source rock samples of the Garau Formation in three other outcrops (F, G, and H). The Van-Krevelen diagram based on elemental analysis data has applied to identify the kerogen type (Peters et al., 2012). As well as, the diagram used to evaluate the thermal maturity of organic matter. The ongoing thermal regime causes the ratios of H/C versus O/C of the kerogen to shift the lower values (Brooks et al., 1987). For the visual kerogen analysis, approximately 50 isolated kerogens have been prepared and palynology has been conducted on five selected samples. Preparing of the kerogen thin sections and palynology was done using the standard method, which is described in the Traverse's work (Traverse, 2007), as follow: first carbonates and silicates of the samples were dissolved by using hydrochloric and hydrofluoric acids, and after neutralization with distilled water, the residue was centrifuged for heavy liquid separation and sieved through a 15-µm mesh size. Permanent mounts were made in glycerin jelly and finally, the kerogen adheres to the lamella. Palynological elements were identified and their percentages were used for sedimentary environment interpretations. Also, the kerogen images have been taken using an optical microscope. Determining the relative proportions of the various organic components contained in the slides, is done through counting analysis. The transmittance colour index (TCI) of the amorphous organic matter has conducted for the thermal maturity assessment.

4. Results and discussion

4.1. Pyrolysis and elemental analysis

The data of hydrogen index (HI; mg HC/g TOC) versus oxygen index (OI; mg $CO_{2/g}$ TOC) (as Table 1) show predominantly kerogen type I and II (Figure 3). On the other hand, the Van-Krevelen diagram based on elemental analysis data (as Table 2) reveals the kerogen type in other way (as Figure 4). The three analyzed data almost are located near the definition boundaries, however, the F location sample has the lowest oxygen content and shifted toward left of the diagram that reveals the properties of type I kerogen. The other two samples (G and H) have type I/II kerogen characteristics. The little O/C ratios or OI indicates severe anoxic conditions in the sedimentary basin of the Garau Formation. In addition, the fewness entrance of clastic sediments into the anoxic carbonate sedimentary environment caused the contents of iron and pyrite has dropped (Table 2) (Hunt, 1996; Hsu and Robinson, 2006; Lewan, 1998; Orr, 1986; Walters, 2006). Under such conditions, the anaerobic bacteria (sulfate reducing bacteria) have altered sulfur contents of the environment. The released sulfur entered the structure of kerogen, rather than react with the surrounding iron to forms pyrite. Then, the expelled hydrocarbon products will be rich in sulfur with high amounts of NSO compounds and asphaltenes rather than saturate and aromatic fractions (Hunt, 1996; Orr, 1986). The Van-Krevelen diagram shows thermal maturity of the oil window (Hunt, 1996; Van Krevelen, 1984).

Table 1

The Rock-Eval pyrolysis and Leco carbon analysis data.

Location name		**TOC (Wt. %)**	**HI (Mg HC/gr TOC)**	**OI (Mg $CO_{2/g}$ TOC)**
A	**Average**	15.9	588.0	8.4
	Maximum	25.7	762.0	25.0
	Minimum	8.1	442.0	2.0

B	**Average**	7.1	449.3	26.8
	Maximum	24.2	621.0	57.0
	Minimum	0.6	213.0	7.0
C	**Average**	16.4	555.2	10.6
	Maximum	19.4	611.0	20.0
	Minimum	10.8	479.0	5.0
D	**Average**	12.4	478.7	39.2
	Maximum	21.4	576.0	258.0
	Minimum	0.3	106.0	4.0
E	**Average**	15.5	497.0	14.6
	Maximum	26.4	667.0	56.0
	Minimum	1.3	93.0	2.0

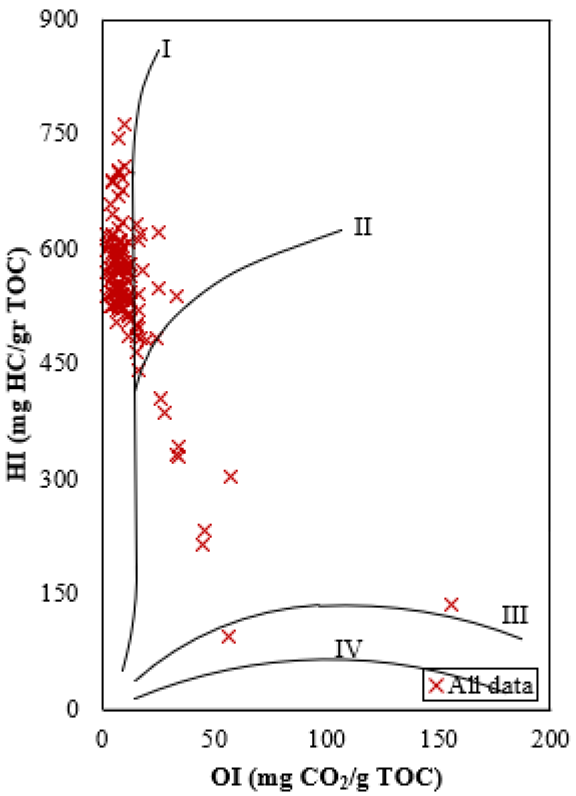

Figure 3

Cross plots of HI versus OI for pyrolyzed Garau Formation rock samples (Hunt, 1996).

Table 2
The percentage of rock sample formers in the Garau Formation rock samples.

Sampling location	C (%)	H (%)	O (%)	N (%)	S (%)	Fe (%)	H/C	O/C
F	73.4	7.1	1.9	0.9	10.7	0.08	1.16	0.02
G	68.4	6.5	3.8	1	10.5	0.7	1.14	0.04
H	67.3	6.3	3.4	1	9	0.03	1.12	0.04
Average	69.7	63.6	3.03	0.97	10.07	0.27	1.14	0.03

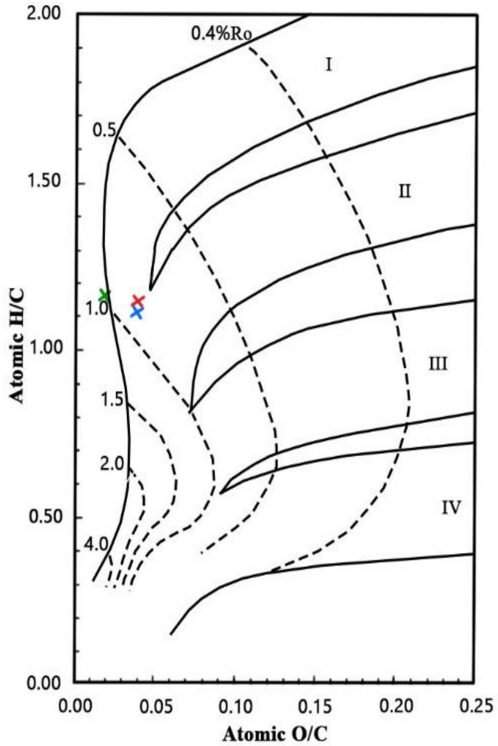

Figure 4
Thermal maturity stage and organic matter type of the Garau Formation (Hunt, 1996).

4.2. Evaluation of thermal maturity

To evaluate thermal maturity, the atomic H/C versus O/C ratio, along with the hydrogen index (HI) versus Tmax have been plotted in figures 4 and 5. Results indicates the immature-early stages of thermal maturation and the beginning of the oil window (Peters and Cassa, 1994; Rimmer et al., 1993; Tissot, and Welte, 1984).

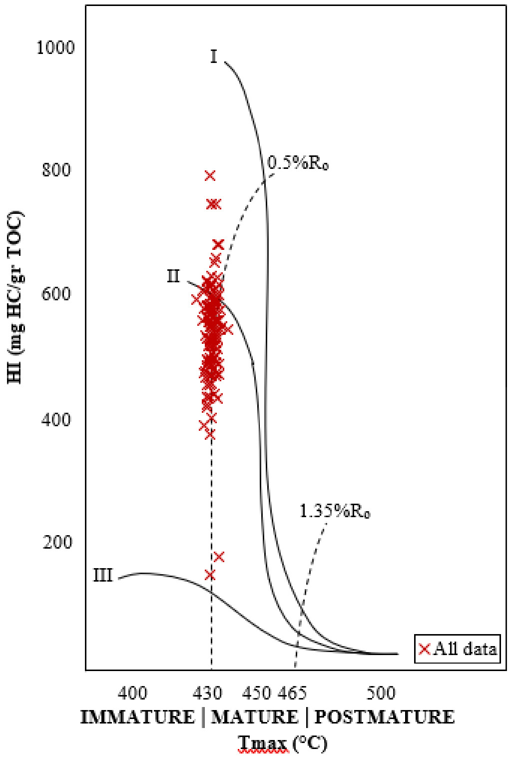

Figure 5

Cross plots of HI versus Tmax, Garau Formation (Rimmer et al., 1993).

The microscopic studies show the existence of some pyrite in the thin sections. Therefore, based on the stoichiometry principles of Skoog et al (Skoog and Leary, 2013), the non-pyritic and pyritic sulfur

contents can be calculated and compared together. The amount of pyritic sulfur contents (Sp) is calculated as the equations 1-3:

$$wt.\%Fe=ma/mFeS_2 \quad (1)$$

$$wt.\%S_2=2\ m_a/mFeS_2 \quad (2)$$

$$Sp= wt.\%S_2\times wt.\%\ Fe\ /\ wt.\%Fe \quad (3)$$

Where, ma is atomic mass, m is molecular mass, wt. %Fe is iron weight percent, wt. %S2 is sulfur weight percent, % Fe is percentage of iron from elemental analysis.

After substituting relevant parameters:

$$wt.\%Fe=46.54,\ wt.\%S_2=53.46,\ Sp._A=0.091\ wt.\%,\ Sp._G=0.804\ wt.\%,\ Sp._H=0.034\ wt.\% \quad (4)$$

Arithmetic and algebraic means are the same and equal to 0.31 wt.%. Since the elemental analysis has already calculated the percentage of total sulfur (S) in the sediments (as Table 2), the difference between mean total sulfur content and mean pyritic sulfur, becomes mean organic sulfur, as; So=S-Sp=10.07-0.31=9.76 wt%. Results was summarized in Table 3.

Table 3
The organic and pyritic sulfur contents in the Garau Formation.

Sampling location	Pyritic sulfur wt.%	Organic sulfur wt.%	TOC wt.%
A	0.091	10.609	11.9
G	0.804	9.696	20.6
H	0.034	8.966	19.6

Overcoming the organic sulfur on pyritic sulfur is an evidence of reduced conditions and suitable for preservation of organic matter from oxidation. The ratio of total sulfur of sediments (S) to organic carbon (C) can be used to recognize anoxic environments from non-marine and freshwater sediments as shown in figure 6 (Berner and Raiswell, 1983; Calvert and Karlin, 1991). Results indicates an obvious distinction with non-marine and freshwater environments. The sedimentation of the Garau Formation has occurred in deep and anoxic waters. Since the kerogen is sulfurized, then, it'll produce hydrocarbon at lower thermal levels rather than similar kerogens at the same conditions. This is because of weaker chemical bonds between carbon and sulfur atoms which is broken easier than others (Mortimer, 1986).

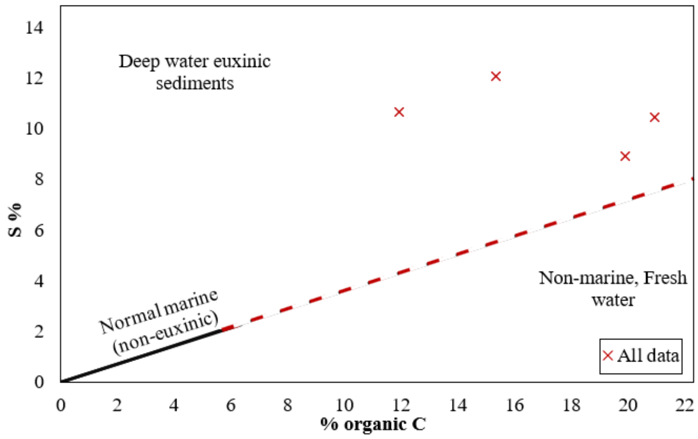

Figure 6
Total sulfur versus organic carbon contents. The dashed line resulted from current study (Berner and Raiswell, 1983).

Based on data plotting of HI versus TOC as shown in Figure 7, the Garau Formation was considered as a good-excellent oil source rock [33]. It should be noted that Figure 7 reveals a remarkable situation; five samples are dropped outside the source quality defining fields. It is difficult to establish a logical relation between the TOC and HI values for these samples due to higher HI values obtained from lower TOC values. Such a situation has caused by in situ bitumen contamination.

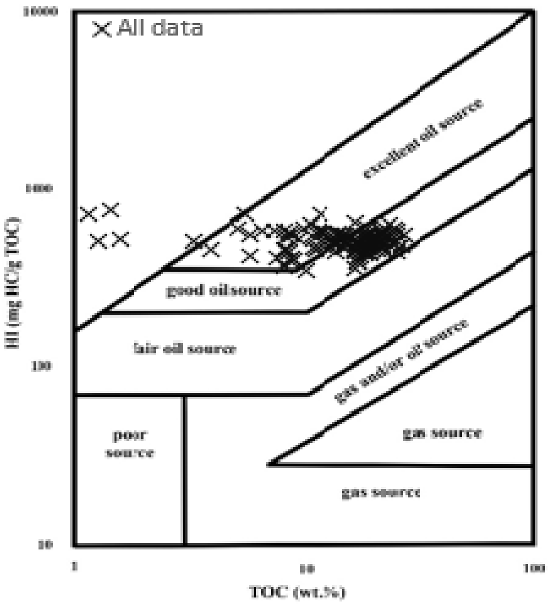

Figure 7
The source rock description based on HI-TOC analysis.

4.3. Visual kerogen analysis and palynology

Organic petrography reveals up to 90% amorphous organic matter (AOM; structure less amorphous products derived from phytoplankton or bacteria and higher plant resins), less phytoclast (fragments of tissues derived from higher plants or fungi), and no palynomorph (organic-walled constituents that remain after acid maceration) in the kerogen thin sections (Mendonça Filho et al., 2011; Tyson, 1993; Vicent, 1995). In addition, slides contain small amounts of frameboidal pyrite (Figure 8).

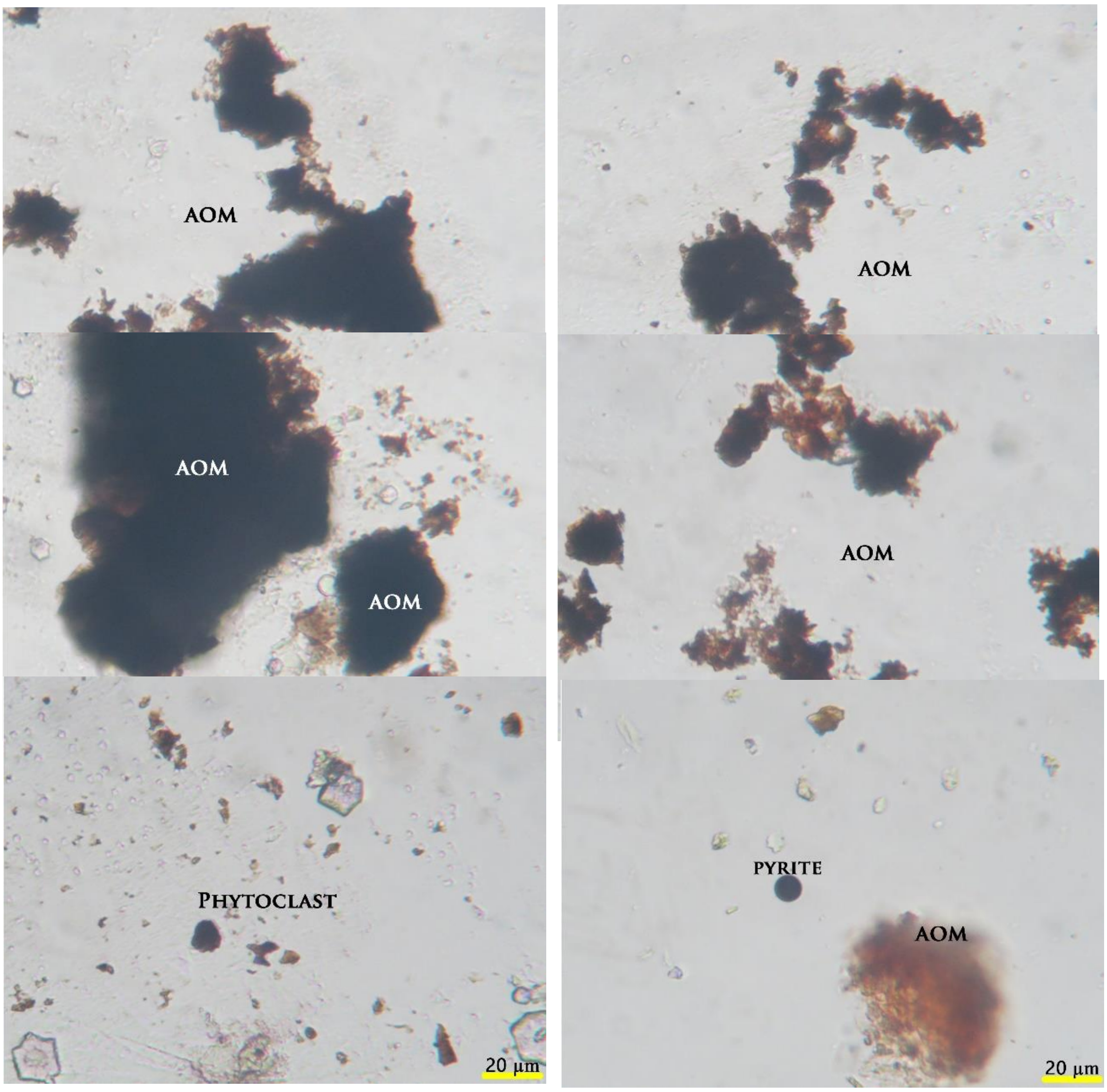

Figure 8
Optical microscopy. Photomicrographs of the Garau Formation include dominantly AOM.

To determine the composition of organic matter and sedimentary environment, the palynological elements data are plotted in a triangular diagram (AAP diagram) as shown in the figure 9 (Suarez-Ruiz, et al., 2012; Tyson, 1995). Where the data is located in IX field, indicates the anoxic condition in the sedimentary basin that is in agreement with the elemental analysis results. Under such conditions, the Garau Formation is formed from AOM-dominate (mostly algal fragments) assemblages, low abundance of palynomorphs, low phytoclasts and spores, low microplankton, and oil-prone kerogens (types I and II) in deep marine, and distal from continental basins by stratified deposits (Tyson, 1995). The organic matter is a mixture of liptinite and exinite-type kerogens (Brooks et al., 1987). As well as, the darkness of AOMs is due to the effect of thermal maturation.

To infer the biological, physical, and chemical processes, the organic facies has assessed. Based on the H/C, HI, and OI data, the Garau Formation has experienced the B and BC organic facies (Tyson, 1995; Jones, 1987). However, due to the low oxygen and high organic carbon contents in the carbonate basin, it's more realistic to classify it in the class of AB or B, which kerogen mainly formed from AOMs, the phytoclasts are low, and the HI is generally between 400 and 650. Although there are some HI values outside this range, the kerogen is generally classified as type I and II. The organic matter has high TOC, and low OI values. The rate of terrestrial matter inputs into the basin was low.

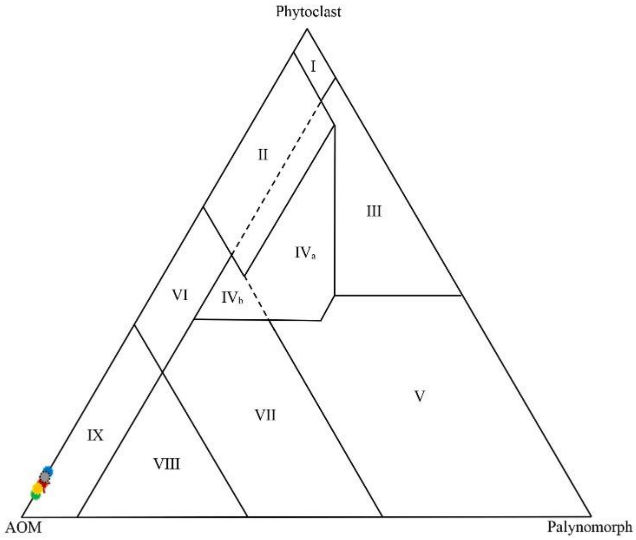

Figure 9

Kerogen plot AOM-Phytoclast-Palynomorph (Tyson, 1995).

The abundance of AOM and organic sulfur is due to happening of a rapid transgression during sedimentation. The sedimentation probably occurred in distal and deep parts of a carbonate platform (Suarez-Ruiz et al., 2012; Tyson, 1995; Batten et al., 1996). The greenhouse effect created by seafloor spreading and volcanic activity during the Middle-late Cretaceous, resulted in the deposition of pelagic organic-rich facies of the Garau Formation during transgression under an oceanic anoxic event (OAE) (Arthur et al., 1985, 1990; Baudin, 2005; Haq et al., 1987; Kobraei et al., 2017; Leckie et al., 2002).

5. Conclusion

The Garau Formation has composed of kerogen type I and II with high contents of sulfur. The quality considered as a good to excellent oil-prone source rock. AOM is the dominant constituent and organic matter is a mixture of liptinite and exinite-type kerogens. The organic facies have distinguished as class of AB and B. Due to anoxic condition and high sulfur contents, along with fewness clastic sediment supply, the sulfur under act of sulfate reduction bacteria has entered the structure of kerogen. The amount of pyritic sulfur (Sp) and organic sulfur (So) contents has calculated, as: Sp= wt. %S2× % Fe / wt. %Fe, and So=S-Sp. Furthermore, based on the high organic sulfur contents of the Garau Formation, it could produce hydrocarbon at lower thermal maturities rather than similar kerogens at the same conditions. The products will be sour and cause some issues, such as corrosion of the pipes.

Nomenclature

OI	Oxygen Index, mg CO_2/g TOC
HI	hydrogen index, mg HC/gr TOC
AOM	Amorphous Organic Matter
S_p	Pyritic Sulfur
TOC	Total Organic Carbon, wt. %
TCI	Transmittance Colour Index
NSO	Nitrogen, Oxygen, Sulfur
ma	atomic mass
m	molecular mass
wt.%Fe	iron weight percent
wt.%S2	sulfur weight percent
%Fe	percentage of iron from elemental analysis
APP diagram	AOM-Phytoclast-Palynomorph plot
OAE	Oceanic Anoxic Event

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