Study of Ferrous to Ferric Ratio in Source Rock Sediments of Jaisalmer Basin of Rajasthan, India, Using Mössbauer Spectroscopy

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Number of sedimentary samples obtained from different depth were studied using ⁵⁷Fe Mössbauer spectroscopy to identify iron phases in samples. These samples were collected from different test wells drilled in Jaisalmer petroliferous basin and also for each well the samples were collected from different depths. Aim of this study was to identify different iron phases and also to obtain information about crucial Fe^{2+}/Fe^{3+} ratio in the organic rich sediments (source rocks). The Fe^{2+}/Fe^{3+} ratio seems to be correlated with the environment of deposition in the sediments. The smaller value of Fe^{2+}/Fe^{3+} ratio indicate poor reducing condition in the sediments which is not favourable for generation of oil/gas in a basin.

Keywords: Mossbauer spectroscopy, Iron phases.

1. INTRODUCTION

Sediments rich in organic matter are known as source rocks [1]. The organic matter can be divided into two broad categories: Humic (derived from terrestrial source) and Sapropelic (derived from marine source). As the time passes, this organic matter is modified by bacteria and undergoes thermal alteration. Most of the organic matter in sediments ultimately changes into insoluble organic matter, termed as 'kerogen'. The Kerogen can be classified in to three types: Type I, II and III. Rocks containing type I and type II kerogen can generate both oil and gas, while those containing type III kerogen, generate dry gases (CH_4 , CO_2 and N_2). Therefore, the nature of kerogen present in sediments is one of the principle factor while determining whether liquid oil, wet gas or dry gas would have been formed during the process of maturation. Furthermore, the physicochemical transformation of organic matter during the geological history of the sedimentary basin cannot be regarded as an isolated process. It is controlled by the same major factors that also determine the variation in the composition of the inorganic solid phase of the sediment; biological activity in the early stage, then temperature and pressure [2]. The mineral matter also includes iron bearing minerals which can be characterized by Mossbauer spectroscopy.

Large number of wells has been drilled in several parts of Jaisalmer basin (India), by drilling agencies like ONGCL (Oil and Natural Gas Corporation India Limited) and OIL, India (Oil India Limited), but economically viable oil has not been explored so far. However many structures have yielded hydrocarbons; mainly methane accompanied with appreciable amount of nitrogen and carbon dioxide for example in Lunar structure

Received: 19.07.11, Accepted: 23.08.11

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(LNR) nitrogen is more than 80 percent while in Dandewala structure (DND) it is less than 40 percent.

In present work sedimentary samples collected from different depths of four test wells namely DND-1 located in Dandewala structure, MNW-1 located in Manewala structure, LNR-1 located in Lunar Structure and BT-3 located in Bhakri-Tibba structure has been studied using ⁵⁷Fe Mössbauer spectroscopy. The Cretaceous and Jurassic sediments are believed to contain source rocks in this basin [3]. The aim of present study is to determine the chemical state of iron in the sedimentary sequences at various depths and to plot the variation of crucial Fe²⁺/Fe³⁺ ratio with depth in these wells. The Fe²⁺/Fe³⁺ ratio plotted as a function of depth seems to be correlated with the redox environment of deposition in sediments which is also among one of the factors favouring the transformation of organic mater into hydrocarbons.

2. EXPERIMENTAL

The Mössbauer absorber were prepared by sandwiching finely ground sediment samples between two paper disc in sample holder (25 mm diameter). All Mössbauer spectra were recorded at room temperature with a conventional constant acceleration Mössbauer spectrometer using a ⁵⁷Co source of 10 mCi initial strength. The Isomer shift has been reported with respect to centroid of a 25 μ m thick α -iron foil spectrum. Details of experimental setup and fitting programme have been already reported [4-10].

3. RESULT AND DISCUSSION

In Figure 1(a) to 1(d), we display some representative Mössbauer spectra recorded at room temperature for samples collected from different depth intervals of wells DND-1, MNW-1, LNR-1 and BT-3 respectively. Depth interval from which the sample is collected has been marked in the figure itself. Majority of the spectra exhibit five doublets marked as AA', BB', CC', DD' and EE' corresponding to iron in pyrite, in siderite, in clay in high spin Fe³⁺ state, clay in high spin Fe²⁺ state and ankerite respectively [11,12]. The relative intensity of doublets varies from sample to sample. For doublet AA^{7} , the Isomer shift (IS) value is centered around 0.30 mms⁻¹ and Quadrupole splitting (QS) value is around 0.60 mms⁻¹. Such IS and QS value are exhibited by both pyrite and Fe³⁺ in some clay minerals. To settle the issue we took the help of acid treatment. It is well known that dilute HNO₃ leaches out pyrite from the samples and have no effect on the clay minerals. The Mössbauer spectra obtained after acid treatment has shown no effect on this doublet AA' indicating that this doublet was not due to pyrite. Hence undoubtedly this doublet AA' is attributed to Fe^{3+} in some clay mineral. For doublet DD' the IS and QS values are centered around 1.10 mms⁻¹ and 2.60 mms⁻¹ respectively corresponding to iron in Fe^{2+} state in clay minerals. Doublet BB' with IS and QS values centered around 1.24 mms⁻¹ and 1.80 mms⁻¹ respectively is attributed to iron in siderite. The doublet EE' with IS and QS values centered around 1.25 mms⁻¹ and 1.65 mms⁻¹ is attributed to iron in carbonate minerals other than siderite [8].





1(d): depths 2395 m

Figure 1: Room temperature Mössbauer spectra of samples collected from different depths (a) DND-1 (2161 m), (b) MNW-1 (3312 m), (c) LNR-1 (1175-1180 m), (d) BT-3 (2395 m).

In Figure 2 to 5, the ferrous to ferric ratio in different wells as a function of depth has been shown. It can be seen that in all these studied wells, this ratio (Fe^{2+}/Fe^{3+}) is relatively smaller (< 1) at the upper depths indicating that in particular sample the relative amount of Fe^{2+} in clay is less in comparison to the relative amount of Fe^{3+} in clay minerals. At the lower depths, in all the studied wells, the ratio (Fe^{2+}/Fe^{3+}) shows an abrupt rise indicating an appreciable increase in the relative amount of Fe^{2+} in clay.



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Figure 2: Distribution of (Fe^{2+}/Fe^{3+}) ratio as a function of depth in well DND-1.

Figure 3: Distribution of (Fe^{2+}/Fe^{3+}) ratio as a function of depth in well MNW-1.



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This appreciable presence of Fe^{2+} in clay minerals at lower depths can be easily explained from argument give by Tissot and Welte [1] according to which at higher temperature two process occurs simultaneously in sediments at one hand hematite is formed due to loss of water from goethite and at the same time the increase in Fe^{2+} in clay minerals take place due to increase in crystallineity of clay present in sediments. It is worthwhile to mention that in some samples of lower depths of these wells iron in oxide (hematite) has been also observed [13] which also indicates towards high temperature condition have prevailed during deposition of sediments in lower depth.

The present study indicates that relatively smaller Fe^{2+}/Fe^{3+} ratio in these studied wells is an indicator of poor reducing environment in the sediments of Jaisalmer basin which is less favourable for generation of oil/gas in a basin.

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