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Application of isotopic analysis results to study origins of CO_2 gases in Malay - Tho Chu Basin

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ABSTRACT

In Malay - Tho Chu basin, CO_2 gases were found with content ranging from few up to 66% by volume. The CO_2 gas amounts increase with depth, CO_2 - rich gases mainly concentrate in I and J sequences from depth of 3000m in the North and 1500m in the South. The gases in this basin could be divided into two groups: Group 1 is hydrocarbon - rich, typically containing 65 - 85% hydrocarbons and less than 20% CO_2; Group 2 is CO_2 - rich, containing 22 - 66% of CO_2 in total gases. The hydrocarbon gases are quite dry because of methane predominance composition and have thermogenic origin. On the other hand, CO_2 gases have both organic and inorganic origins. The organic CO_2 gases showing low - CO_2 content are characterized by a lighter $\delta_{13}C_{CO2}$ varying from - 17.8‰ to - 10.8‰. The inorganic CO_2 gases make up high content and occur at deeper reservoirs. These CO_2 gases are isotopically heavy with $\delta_{13}C_{CO2}$ values from - 9.3‰ to 0.5‰. The large amount of CO_2 gases in Malay - Tho Chu basin is believed to be generated from the mineral thermal decomposition which occurred in the deep troughs as Tho Chu graben and central trough of Malay basin with depth of over 14km.

1. Introduction

In continental shelf of Vietnam, Malay - Tho Chu basin located in the Southwest (Figure 1). Over the past 40 years, petroleum exploration has been conducted and commercial oil and gas fields have been discovered such as Cai Nuoc oil - gas field and Ac Quy, Kim Long, Ca Voi gas fields. However, beside the hydrocarbon accumulations, some wells have been encountered the CO_2 gas with remarkable content of 20% - 66% (Figure 1), which has hindered the further petroleum exploration activities in this area. The objectives of this study are to understand and document the different characteristics, origins and natural processes involved in all types of occurrence of subsurface and near - surface CO_2, and then vestigate the origins and accumulation of CO_2 gases in Malay - Tho Chu basin.

2. Geology setting

The geological and structural characteristics of Malay - Tho Chu basin are typically similar to those of the other sedimentary basins in Vietnam. It is made up of two main stratigraphic units as

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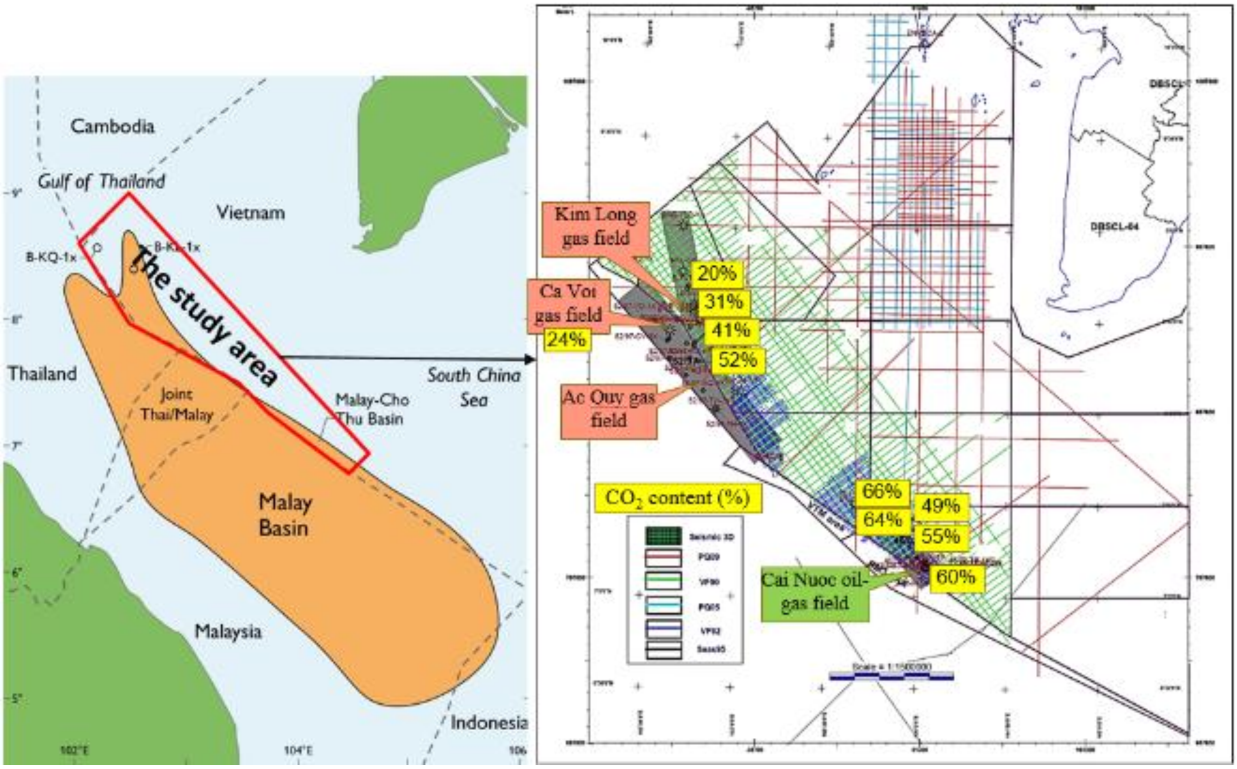

Figure 1. The location of Malay - Tho Chu basin in Gulf of Thailand area and the encountered CO_2 gas content at wells in this basin within continental margin of Vietnam

followings:

Pre - Tertiary and Tertiary (Phan Trung Dien et al, 1995; Truong Minh et al., 1997). The Pre - Tertiary unit was formed by various phases during the pre - rifting period. The formation is of heterogeneous lithology, of different ages in different basins and is composed of consolidated basement complex, carbonate, and intrusive and extrusive rocks of the Paleozoic and Mesozoic. The Tertiary unit is the Paleogene - Neogene - Quaternary sedimentary sequence, directly overlying the Pre - Tertiary basement. The Tertiary geologic history and evolution of the Malay - Tho Chu basin can be divided into the following main stages:

2.1. Eocene (?) - Oligocene rifting stage

The most influential tectonic activity in the study area is the intra - cratonic rifting process (Phung Si Tai et al., 2001;) which is often referred to as the synrift period that formed major Tertiary sedimentary basins in the Malay - Tho Chu basin and Pattini trough (Figure 2). Sediments were at first separated by half - grabens. These sub - basins were later filled with sediments of continental - lacustrine, delta, and coastal marine facies. These deposits are mainly sand, shale, fluvio - lacustrine and braided stream deposits with the oldest being Oligocene sediments. As faults initiated from the Pre - Cenozoic basement, Oligocene formations are often differentiated, separating the paleo - topography into heterogeneous Pre - Cenozoic basement highs and lows, and creating very a complex geological framework. In the end of the Oligocene, due to an up - lift event, sedimentation was interrupted and eroded, marked by the Upper Oligocene - Early Miocene unconformity.

2.2. Miocene - Quaternary stage (Post - rift)

The Early Miocene period started with subduction and sagging - marine transgression phases, typical activities characterizing a transitional period from syn - rift to post - rift (Phung Si Tai et al., 2001; Petronas, 1999) (Figure 2). During the Middle Miocene, basin subsidence continued in response to geothermal cooling of the lithosphere. The period from the Late Miocene

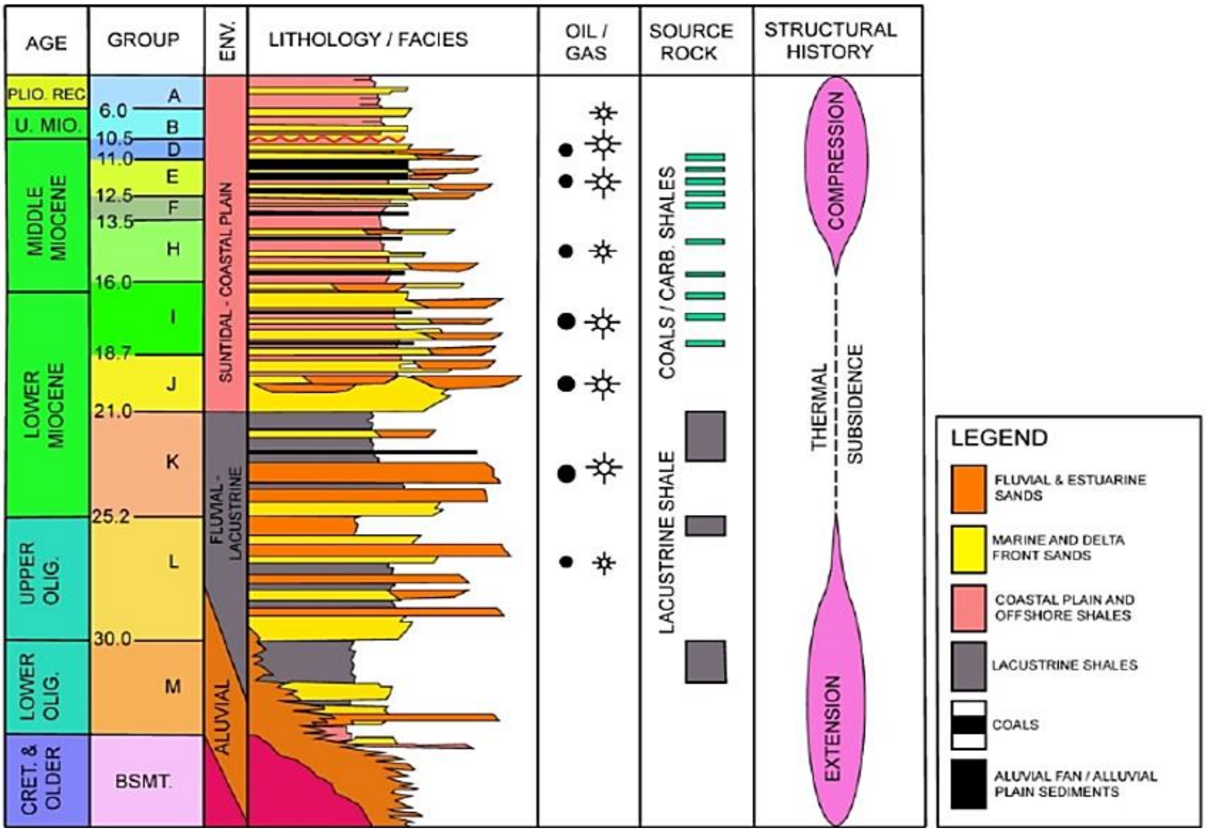

Figure 2. Summarized stratigraphic column of the Malay - Tho Chu basin (After LML, 1998; Petronas, 1999; Gilmont et al., 2001; Truong Son JOC, 2003).
Legend: 1. Fluvial and estuary sand; 2. Marine and deltafront sand; 3. Aluvial fan/aluvial plain; 4. Carbonate; 5. Coastal plain and marine clay; 6. Lacustrine clay; 7. Volcanic; 8. Coal.

to present day is the latest phase of the basin development process, which is the continuation of the post - rift period. In the Pliocene - Quaternary, subsidence slowed down and became stable, while wide - spread marine transgression prevailed. Basins and adjacent sub - basins in the Gulf of Thailand were connected. Sedimentary cover was mostly horizontal and unaffected by faulting or folding activities, forming the current structural pattern.

3. Database

The analysis results of gas samples were taken from done geochemical reports provided by various service companies. According to this, ninety gas samples were collected during drill stem tests (DST) and module formation tests (MDTs) at wells of Malay - Tho Chu basin. The gases were analyzed for their chemical compositions, stable carbon isotopes ($\delta^{13}C$) of C_1, C_2, C_3 and CO_2. In this paper, in order to elucidate the differences in distribution and origins of CO_2 gases through the whole basin, the results were discussed for the Northern and Southern parts of the basin.

4. Methodology: formation of CO_2 gases in sedimentary basins and isotope study

4.1. CO_2 formation in sedimentary basins

In many sedimentary basins over the world, CO_2 gas is found in both reservoirs and surface samples. Sources of CO_2 gas are both organic and inorganic (Figure 3). The organic CO_2 sources include microbial action, kerogen breakdown and in reservoir reactions. Inorganic CO_2 comes from deep sources of carbonate decomposition, volcanic activities and metamorphic fluids.

4.1.1. Kerogen breakdown

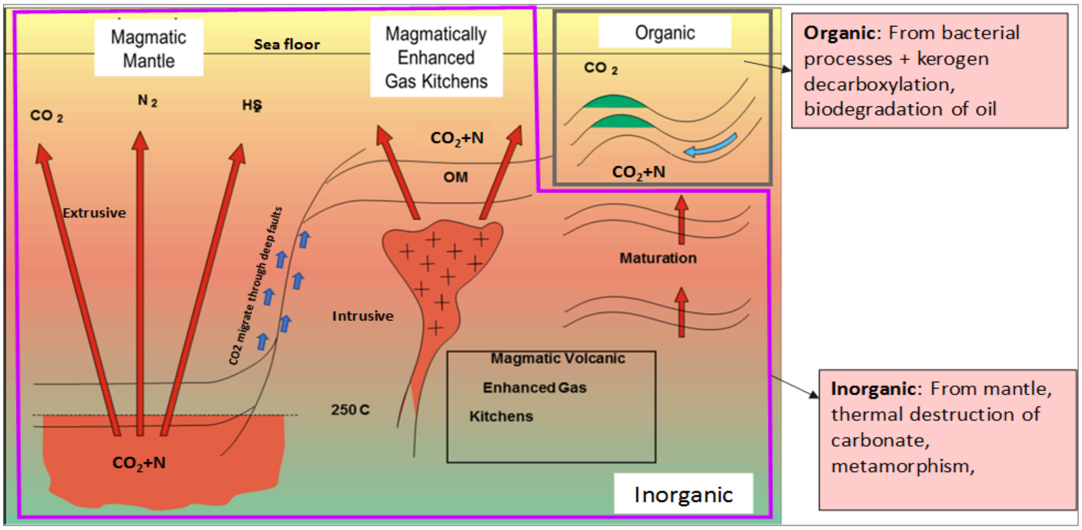

Figure 3. Diverse origins of CO_2 gas (Schoell, 2013).

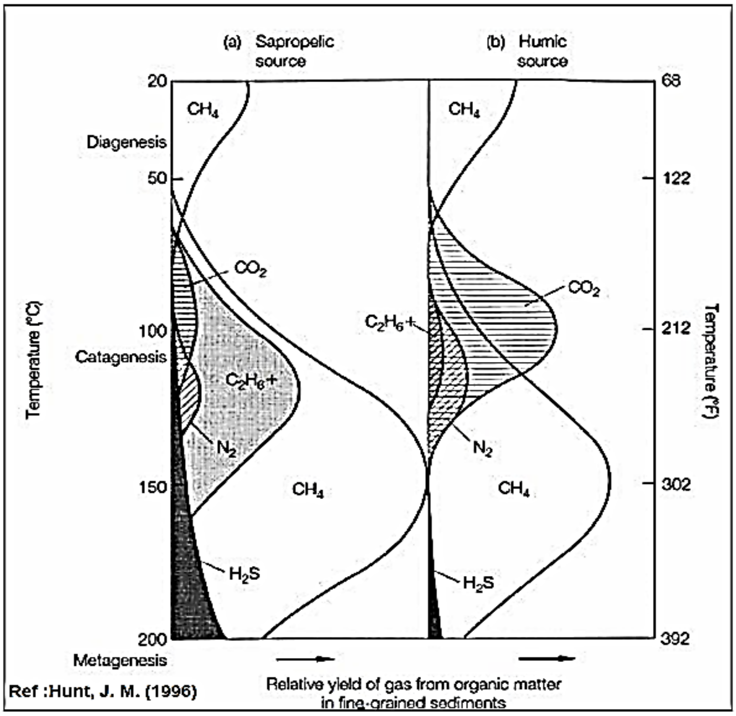

Figure 4. The generation of gases from organic matter with temperature. The C_2^+ represents hydrocarbon gases heavier than methane. The N_2 is generated initially as NH_3. Inorganic sources of CO_2, H_2S and N_2 are not shown (Hunt, 1996).

Organic CO_2 gas can be derived from the thermal decarboxylation of organic matter which occurs during diagenesis and catagenesis and is mostly completed at the end of the oil - window. Continental derived humic materials contribute most of the organic source CO_2 (Hunt, 1996) (Figure 4).

4.1.2. Microbial action

In biogenic gas system, organic matter is buried beyond the sulfate reduction zone, therefore organic CO_2 gas can also be generated by methanogenic bacteria at temperature up to about 70°C (Figure 5). Two main metabolic

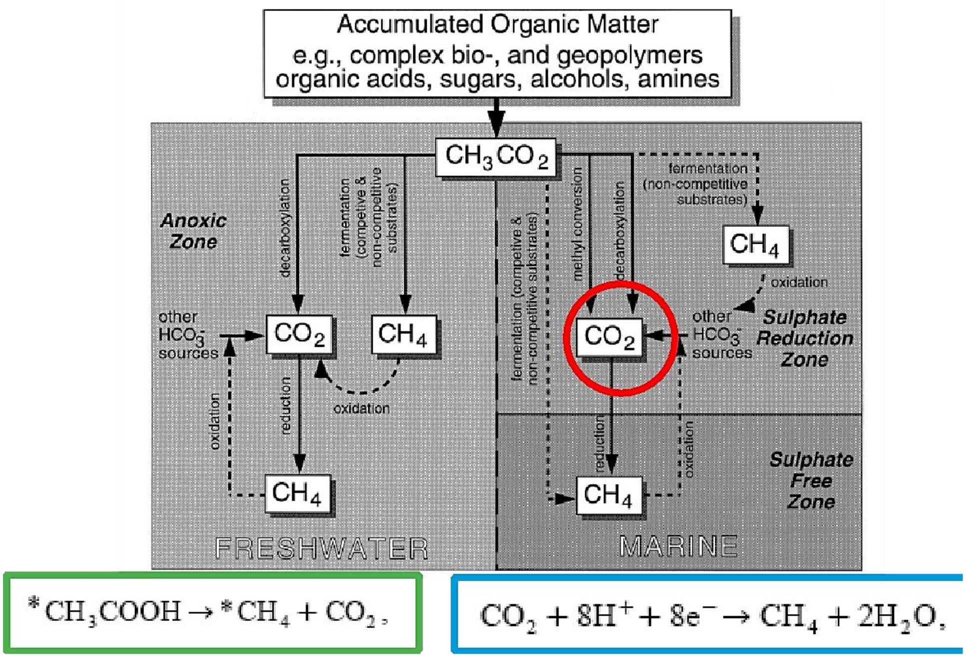

Figure 5. Schematic diagram showing methanogenic pathways in marine and terrestrial environments (Marty, 1992).

pathways are involved. Acetate fermentation is a disproportionation reaction which yields both CO_2 and CH_4:

$$CH_3COOH \rightarrow CO_2 + CH_4$$

Carbon dioxide reduction uses dissolved hydrogen, itself a by - product of bacterial activity, as a reducing agent:

$$CO_2 + 8H^+ + 8e^- \rightarrow CH_4 + 2H_2O$$

4.1.3. Carbonate decomposition

When high temperature magma penetrates carbonate rocks, the carbonate decomposes and gives off CO_2 (Hunt, 1996) as follows: $CaCO_3 \rightarrow CaO + CO_2$.

The CO_2 migrates as a gas and can be trapped in reservoir rocks of nearby structures. The typically temperatures are around 840 - 850°C. Besides, in sedimentary basins where iron or magnesium carbonates are present, CO_2 also is generated from silicate carbonate reactions. The Figure 6 shows the point at which a separate CO_2 - rich gas phase can be produced in sedimentary basins if the sediments reach temperatures of approximately 330°C (Cathles and Schoell 2007). The CO_2 titration and carbonate precipitation cannot occur in the reservoirs but must rather occur along the migration pathways (Cathles and Schoell 2007) (Figure 7).

4.1.4. Volcanic activities and metamorphic fluids

Along with H_2O, CO_2 is one of the lightest volatiles (materials of relatively low melting point), found in the mantle (Wilson, 1989). The fluid nature of the asthenosphere, or upper mantle of the earth, ensures that lighter volatiles are fractionated, buoyed towards the surface, and either extruded or outgassed into the atmosphere via volcanoes and faults.

4.1.5. In reservoir reactions

At temperatures up to about 80°C, petroleum in subsurface reservoirs is often biologically degraded (Head et al.,2003). The Figure 8 shows the putative chemistry of hydrocarbon degradation in most petroleum reservoirs with an absence of abundant sulphate. The overall conversion of hydrocarbons to biomass, methane and carbon dioxide may well involve water - hydrocarbon reactions (Zengler et al.,1999; Widdel and Rabus, 2001) with the carbon dioxide produced being further reduced to methane using hydrogen produced either externally to or within the reservoir.

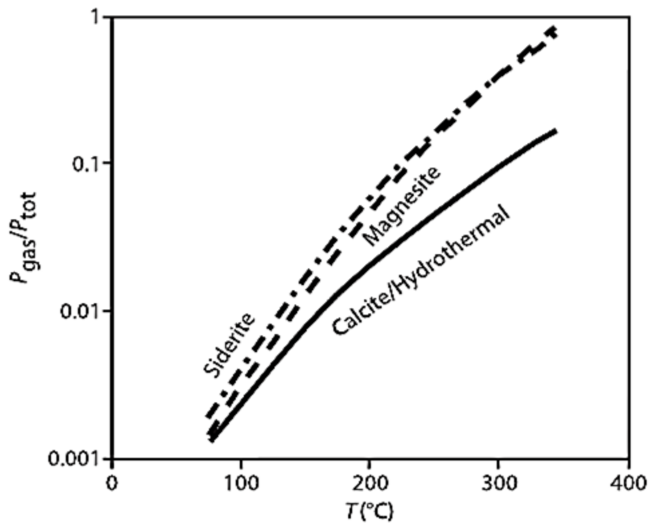

Figure 6. Gas pressure (P_{CO2} + P_{steam}) divided by pore fluid pressure (P_{tot}) calculated for the calcite, siderite and magnesite buffers as a function of temperature with pressure determined from the S&E P - T trajectory. When the gas to total pressure ratio reaches 1, a separate CO_2 - rich gas phase will expel from the pore fluids. High mole fraction CO_2 are produced where basin sediments with siderite or magnesite are heated to temperature approximately >330°C (Cathles and Schoell 2007).

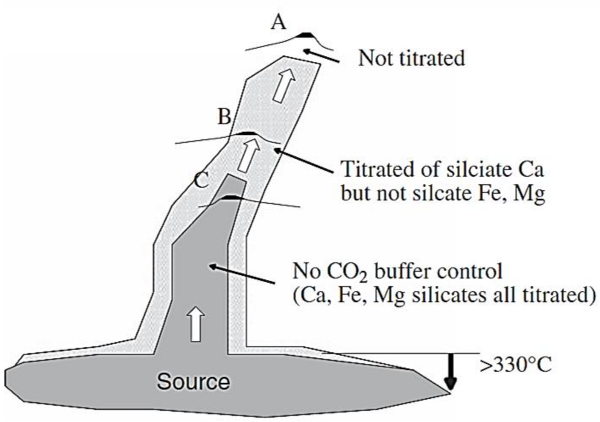

Figure 7. Conceptual scheme of CO_2 generation and titration during migration (Cathles and Schoell, 2007).

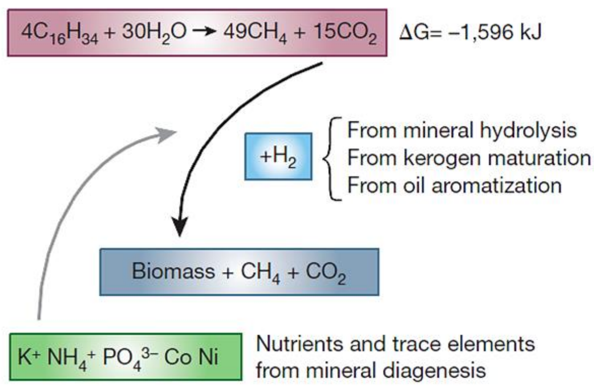

Figure 8. The putative chemistry of hydrocarbon degradation in most petroleum reservoirs with an absence of abundant sulphate (Head, et al., 2003).

4.2. Isotope approach

In biosphere, there are three carbon isotopes containing 6, 7 and 8 neutrons giving them atomic masses, respectively of 12, 13 and 14 (Hunt, 1996). The isotopes ^{12}C and ^{13}C are stable, but ^{14}C decays to ^{14}N. The isotope ^{13}C is distributed through sediments of all geological ages, thus, it can solve many geochemical problems because its difference in mass relative to ^{12}C results in fractionation by both biological and physical processes. The standard that has been used most widely in the literature over the years is a belemnite from the Peedee Formation in South Carolina (PDB). The Table 1 shows a composition of the actual ^{13}C content as a percentage of total carbon for three materials.

The contents of ^{13}C in various materials are different because of kinetic isotope effect, equilibrium isotope effect, dilution effect and conservation of isotope. The different sources of carbon dioxide cause different $\delta^{13}C$ values of CO_2, as shown in Figure 9.

Table 1. Variation in Carbon - 13 in natural materials.

	Percent of carbon - 13	δ 13C‰ relative to PDB
Peedee belemnite (PDB)	1.1112	0
A typical limestone	1.1162	+5
Plankton lipids	1.0862	- 25

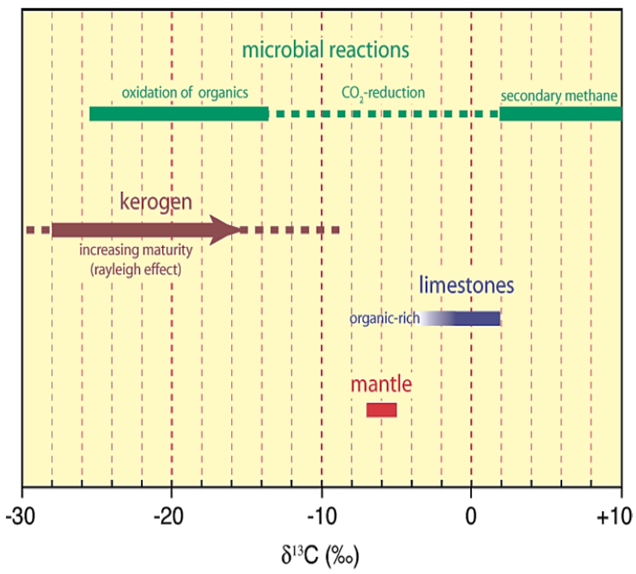

Figure 9. Different origins of CO_2 gases based on carbon isotope values.

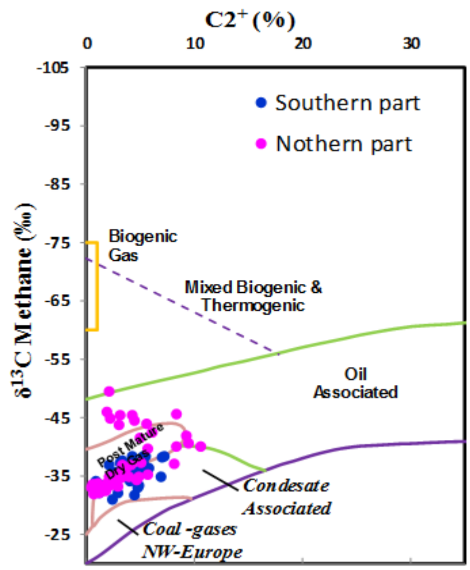

Figure 10. The plot of $\delta^{13}C$ methane vs. C_2 content (% volume).

5. Results and discussion: Origins of CO_2 gases in Malay - Tho Chu basin

5.1. Hydrocarbon gases

In the study area, the gas reservoirs were encountered in Early to Middle Miocene sandstones (*J*, *I*, *H* and F sequences). In the Northern part, depths of gas reservoirs vary in a wide range from 1700 - 3600m while in the Southern part, reservoirs are shallower with depths of 1400 - 2300m. The gases collected from both Northern and Southern parts are methane - rich gases with C1/C1 - 5 ratios by volume ranging from 0.70 to 0.99. Based on the stable carbon isotope of methane, ethane, C1/C1 - 5 ratio and non - hydrocarbon gas concentration, natural gases in the study area is believed to have thermogenic origin.

These gases have quite light $\delta^{13}C_1$ values of - 45‰ to - 31‰. On the $\delta^{13}C_1$ vs. C_2 plot, gases fall into oil - associated category (Figure 10).

The $\delta^{13}C_2$ values are from - 32.24‰ to - 20.59‰. The relationship diagram among $\delta^{13}C$ of methane, ethane and vitrinite reflectance (%*Ro*) shows that these gases were generated from high mature source rocks in main oil generation to wet gas & condensate phase (gases from shallower reservoirs) and dry gas phase (gases from deeper reservoirs) (Figure 11.a). Moreover, many gases in the Northern part located in coal - type thermogenic gas category (Figure 11.b) suggests that these gases related to source rocks containing dominantly humic organic matter deposited in a deltaic setting.

5.2. The origins of CO_2 gases

Gases were discovered mostly in the Northern part of Malay - Tho Chu basin with the main produced gas fields namely Ca Voi, Kim Long and Ac Quy. In the Southern part, both oil and gas were found. The gases composed of variable amounts of CH_4 relative to CO_2 and can be divided into two groups: Group 1 is hydrocarbon - rich, typically containing 65 - 85% hydrocarbons and less than 20% CO_2; Group 2 is CO_2 - rich, containing 22 - 67% of CO_2 in total gases (Figure 12.a).

The CO_2 content (% volume) of all gas samples are plotted on diagrams with depth for each sequence which illustrates different reservoirs. As mentioned above, the depth of reservoirs in the North varies wider than that in the South. In the North, the CO_2 - rich group mainly encountered in J sequence from deeper than 3000m (Figure 12.b). In the South, the high CO_2 content group are encountered at shallower depth of 1500 - 2500m and also concentrated in *I* and *J* sequences (Figure 12.c).

The carbon isotopic composition of CO_2 has been widely used as a diagnostic indicator of its origin. Organic CO_2 derived from kerogen decarboxylation is characterized by $\delta^{13}C_{CO2}$ lighter than - 10‰ (Hunt, 1979; Andresen et al., 1994; Dai et al. 1996). In contrast, the inorganic CO_2 is isotopically heavier than -8‰. The $\delta^{13}C_{CO2}$ values are typically in the range of -8‰ to -2‰ for the mantle and magmatic CO_2 gases (Pineau et al., 1983; Cornides, 1993; Dai et al., 1996; Marty and Tolstikhin, 1998), and from -3.5‰ to +3.5‰ for the CO_2 gases related to thermal metamorphism of carbonate rocks (Clayton et al., 1990; Dai et al. 1996, Zhang et al., 2008). CO_2 gases in the study area have carbon isotope values ranging from -17.8‰ to 0.5‰, therefore, CO_2 gases have both organic and inorganic origins (Figure 13).

The organic CO_2 gases belong to Group 1 in which CO_2 content is lower than 20% volume of total gases (Figure 13). The organic CO_2 gases are characterized by a lighter $\delta^{13}C_{CO2}$ ranging from -

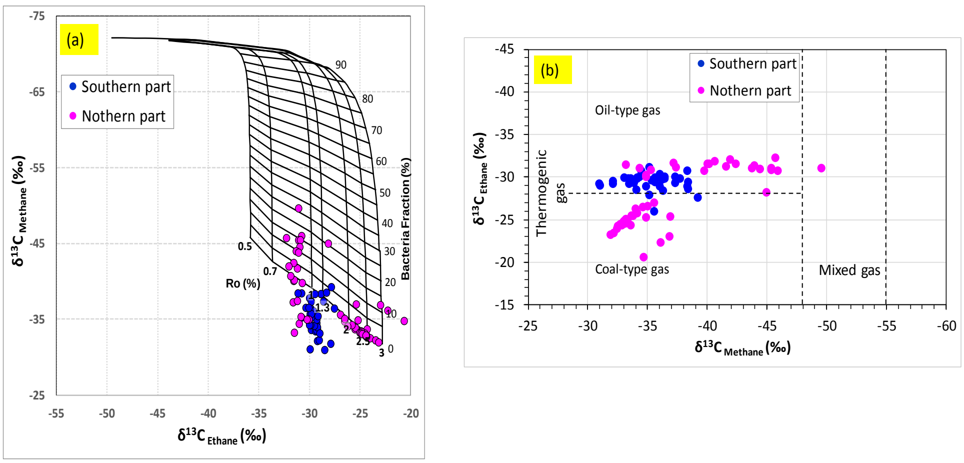

Figure 11.(a,b) Carbon isotope values of methane and ethane of gas samples in the study area.

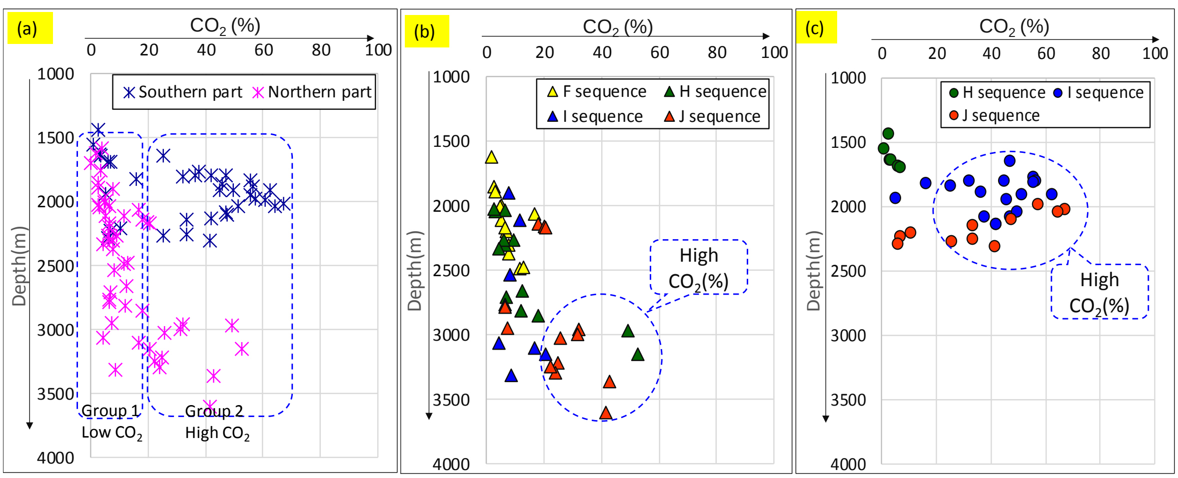

Figure 12. The CO_2 content (%) with depth. (a) In the study area. (b) The Northern part. (c) The Southern part.

14.4‰ to - 10.8‰ in the North and from - 17.8‰ to - 11.9‰ in the South (Figures 13, 15a&b). As mentioned above, all hydrocarbon gases in the study area have thermogenic origin, therefore, the organic CO_2 gases are believed to not related to biogenic activities. During thermal maturation of organic matter, only a small quantity of CO_2 gas can be generated (Hunt, 1996). Thus, the CO_2 gases generated from organic matter could not distribute significantly to total volume gases. In the North, organic CO_2 gases come from shallower reservoirs in *F* and *H* sequences (Figure 14). In the South, CO_2 gases in *H* sequence and some in *I* and *J* sequences have organic origin (Figure 14).

The inorganic CO_2 gases belonging to both Group 1 and Group 2 show various CO_2 contents from low to extremely high (Figure 13). The inorganic CO_2 gases occur at deeper reservoirs, almost in I and J sequences in both the North and the South (Figure 14). This trend is clearer in the North. These CO_2 gases are isotopically heavy with $\delta^{13}C_{CO2}$ values from - 9.3‰ to 0.5‰ (Figure 15a&b). The wide range changes of $\delta^{13}C_{CO2}$ values suggest that these inorganic CO_2 gases could be derived from volcanic/magmatic activities or the mineral thermal decomposition (Figure 15a&b).

However, the origins of CO_2 gases in basins cannot be resolved by their $\delta^{13}C_{CO2}$ values alone because they are not unique and an overlap exists for the magmatic and crustal inorganic CO_2 gases. Mantle fluids are characterized by higher $^3He/^4He$ ratios than those of atmosphere (Ra = 1.4x10^{-6}). Therefore, the fluids derived from the crust show *R/Ra* ratios less than 1 (mostly <0.2; Mamyrin and Tolstikin, 1984) and the volcanic/magmatic and alteration of carbonate origins can be distinguished by isotope values of rare gases. Unfortunately, in the study area, the isotope values of mentioned rare gases are not available.

In the Malay basin, magmatic activities occurred mainly during Triassic to Late Cretaceous (Tjia, 2000) and were nearly not observed in Tertiary. Therefore, it is believed that the large amount of CO_2 gases could be generated from the mineral thermal decomposition source. The Vietnamese part of the Malay basin comprises a large and deep Paleogene pull - apart basin formed through Middle or Late Eocene to Oligocene left - lateral strike - slip along NNW - trending fault zone (Fyhn et al., 2010). In the North, the deepest area in Tho Chu graben is over 10km. The maturity model of a section across the Malay trough to block 46 in the Southwest - Northeast direction shows that sediments reach 300°C from deeper than 8km where the mineral decomposition reactions can happen (Figure 16). Consequently, the deep troughs as Tho Chu graben and central trough of Malay basin with depth of over 14km (Mansor et al., 2014) are believed to be the kitchen where calcareous sediments were deeply buried and decomposed to form very large volumes of CO_2 gas (Figure 17).

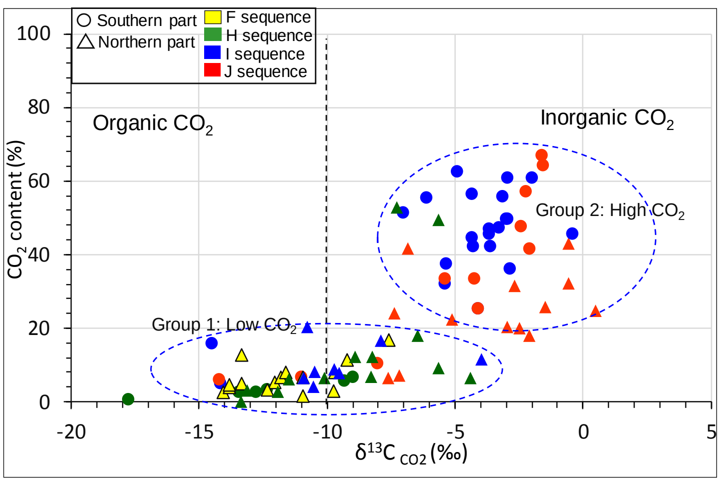

Figure 13. The $\delta^{13}C$ values vs. content (%) of CO_2 gases.

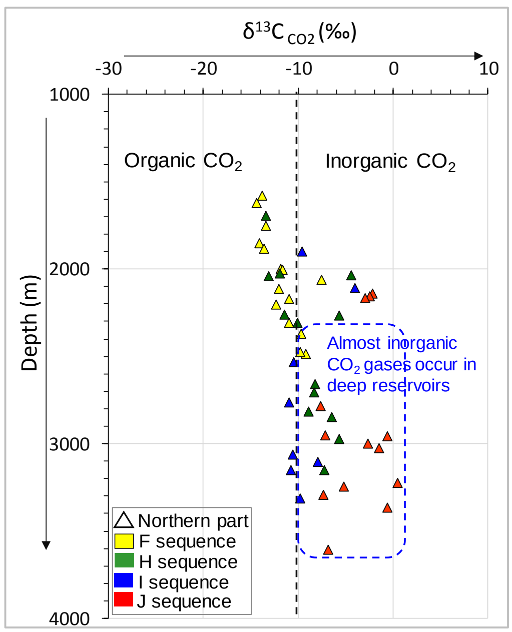

Figure 14. The $\delta^{13}C$ values with depth.

6. Conclusion

- In both the Northern and Southern produced gas fields and oil - gas fields within the study area, CO_2 gases were encountered with percentage ranging from 5% to 66% volume of total gases.

- The hydrocarbon gases in the study area are methane - rich gases and have thermogenic origin.

- The discovered gases composed of variable amounts of CH_4 relative to CO_2 and can be divided into two groups: Group 1 is hydrocarbon - rich, typically containing 65 - 85% hydrocarbons and less than 20% CO_2; Group 2 is CO_2 - rich, containing 22 - 66% of CO_2 in total gases.

- In the Northern part, the CO_2 - rich group mainly encountered in J sequence from deeper

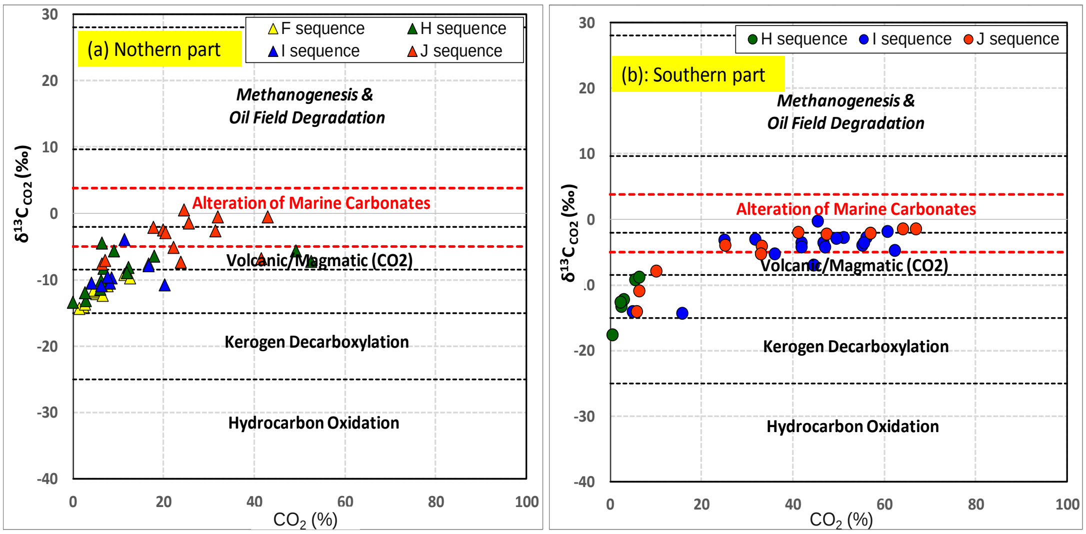

Figure 15. (a,b) The origins of CO_2 gases based on $\delta^{13}C_{CO2}$.

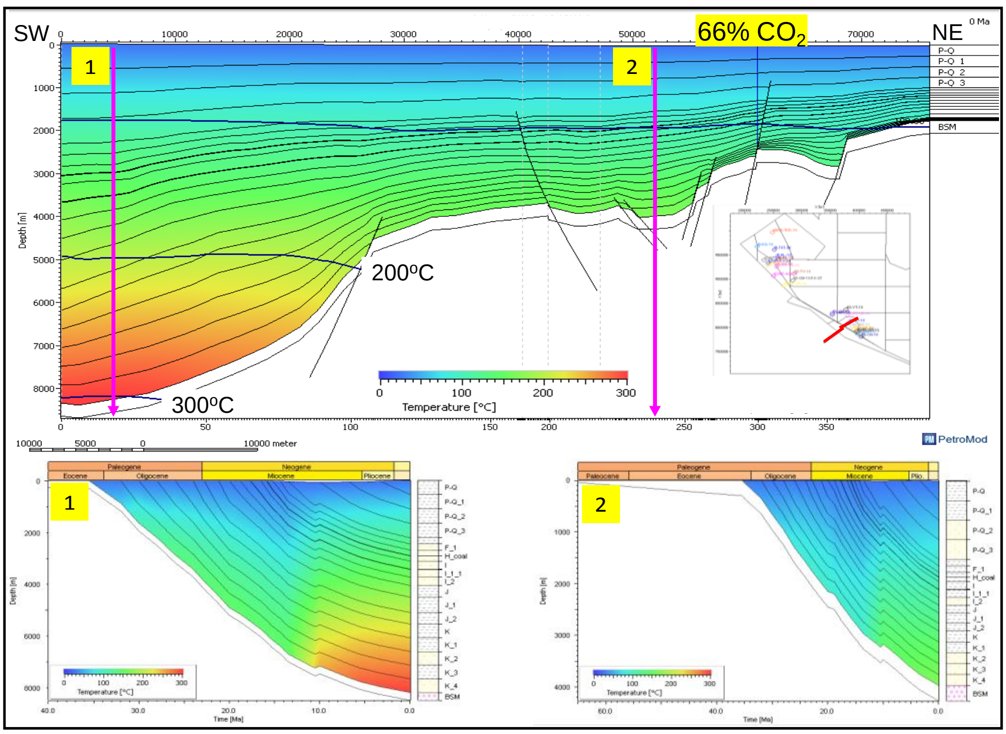

Figure 16. The maturity model of a section across the Malay trough to the Southern part of the study area in the Southwest - Northeast direction (Talisman - 2014).

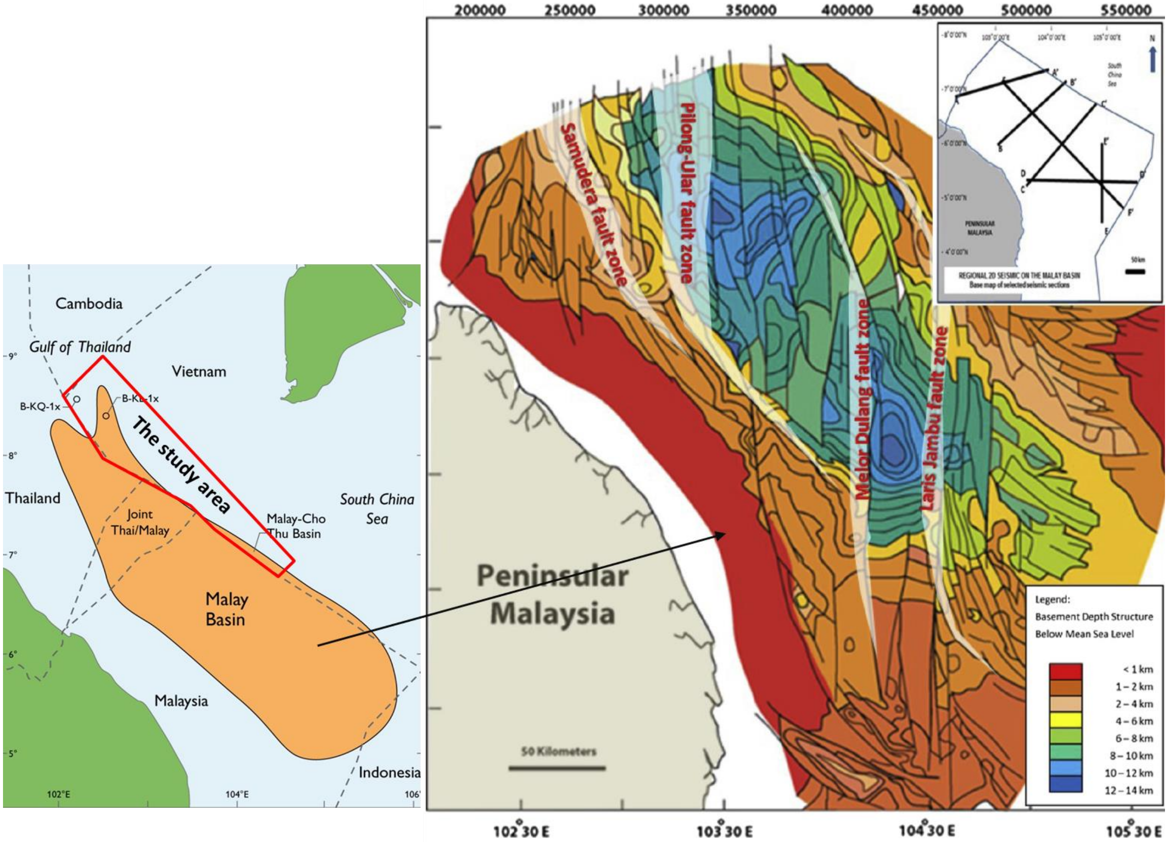

Figure 17. The depth structural map of basement of Malay basin, modified after Musbah et al., (unpublished 2005), with projected maximum depths of some 14km below mean sea level using seismic velocities (Mansor et al., 2014).

than 3000m. In the Southern part, the CO_2 - rich group are encountered at shallower depth of 1500 - 2500m and also concentrated in I and J sequences.

- CO_2 gases in the study area have both organic and inorganic origins. The organic CO_2 gases belonging to low - CO_2 group (<20% CO_2) are characterized by a lighter $\delta^{13}C_{CO2}$ ranging from - 17.8‰ to - 10.8‰. The inorganic CO_2 - rich gases occur at deeper reservoirs, almost in I and J sequences. These CO_2 gases are isotopically heavy with $\delta^{13}C_{CO2}$ values from - 9.3‰ to 0.5‰. The large amount of CO_2 gases in Malay - Tho Chu basin could be generated from the mineral thermal decomposition which occurred in the deep troughs as Tho Chu graben and central trough of Malay basin with depth of over 14km.

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