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Paleogene sediment complex Geological factors impacting on its RESERVOIR QUALITY AND HYDROCARBON POTENTIAL **Editor-in-chief** Dr. Sc. Phung Dinh Thuc

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Management Vietnam Petroleum Institute

Contact Address

16th Floor, VPI Tower, Trung Kinh Street, Yen Hoa Ward, Cau Giay District, Ha Noi Tel: (+84-04) 37727108 Fax: (+84-04) 37727107 Email: tapchidk@vpi.pvn.vn Mobile: 0982288671

Designed by

Le Hong Van

Cover photo: . A geological structure in Vinh Hy, Ninh Thuan Province. Photo: Le Khoa







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Prime Minister Nguyen Tan Dung highlighted the importance of Nghi Son Refinery and Petrochemical Complex. Photo: Nhat Bac



Nghi Son Refinery and Petrochemical Complex Project breaks ground

Nghi Son Refinery and Petrochemical Complex Project with a capacity of 10 million tons/year is the largest-ever project to be built in Vietnam with the most modern technology. Speaking at the groundbreaking ceremony of the project in Thanh Hoa Province on 23 October 2013, Prime Minister Nguyen Tan Dung emphasised that this project will go into operation in 2017 and together with Dung Quat Refinery will meet two-thirds of the domestic demand for petroleum products.

On 23 October 2013, Nghi Son Refinery and Petrochemical L.L.C. (NSRP), a joint venture between the Vietnam National Oil and Gas Group (Petrovietnam), Kuwait Petroleum International (KPI/KPE), Idemitsu Kosan Co., Ltd. (IKC) and Mitsui Chemicals Inc. (MCI) launched the groundbreaking ceremony for Nghi Son Refinery and Petrochemical Complex in Thanh Hoa province.

With the largest foreign direct investment (FDI) in Vietnam so far (over 9 billion USD), the Nghi Son Refinery and Petrochemical Complex will be an important factor to attract further foreign investors into Vietnam, especially investors from Japan and Kuwait. Built on 400ha at Nghi Son Economic Zone (Thanh Hoa province), the Nghi Son Refinery and Petrochemical Complex will robustly boost the socio-economic development of the South of Thanh Hoa province, the North of Nghe An province and the vicinities. The project is expected to create jobs for tens of thousands of people during the construction phase and for thousands of people during the operational phase. The Complex is scheduled to start commercial operation in 2017 with a refining capacity of 200,000 barrels/day (equivalent to 10 million tons/year), and will supply about 40% of Vietnam's petroleum products demand in 2017.

Speaking at the groundbreaking ceremony, Prime Minister Nguyen Tan Dung stated that the Nghi Son Refinery and Petrochemical Complex is a project of very important significance to the socio-economic development and the assurance of energy security for the nation. According to its schedule, the project will go into operation in 2017 and together with Dung Quat Refinery will meet two-thirds of the domestic demand for petroleum products.

The Prime Minister praised the efforts made by Petrovietnam, the foreign investors, and the Nghi Son Refinery and Petrochemical L.L.C. to complete a very large workload in a short time. At the same time, the Prime Minister expressed his appreciation of the efforts made by the Party Committee, the local authorities, and the people of Thanh Hoa province to relocate thousands of

PETROVIETNAM



Prime Minister Nguyen Tan Dung and delegates announced the groundbreaking ceremony of Nghi Son Refinery and Petrochemical Complex Project. Photo: CTV

households in the new residential areas to make the site available for the project.

The Prime Minister asked the investment owners and NSRP as well as the EPC contractors to maximise their expertise and capacity to complete the Nghi Son Refinery and Petrochemical Complex Project in 40 months, making sure that it will operate safely with high quality and efficiency. The Prime Minister also asked the relevant ministries of Vietnam, especially the Minister of Industry and Trade of Vietnam, to co-ordinate and co-operate with Thanh Hoa province and the investment owners to remove arising difficulties and obstacles, closely monitor and promptly provide assistance to help this project go as scheduled.

Kuwaiti Deputy Prime Minister and Minister of Oil Mustafa Al- Shemali said: "The Nghi Son Refinery and Petrochemical Complex Project is of strategic importance because it is a symbol of solidarity between the people of 3 countries Kuwait, Vietnam and Japan. The project is the crystallisation of common resources, experience and commitment, which are fundamental to the long-lasting co-operation in the future between the three countries not only in the oil industry but also in all other areas".

According to Dr.Sc. Phung Dinh Thuc, Chairman of Petrovietnam, in implementing the development strategy of Vietnam's oil and gas sector, Petrovietnam in recent years has vigorously expanded its activities to the refining and petrochemical sector, whilst promoting oil and gas exploration and production at the same time. The completion of the Dung Quat Refinery with a capacity of 6.5 million tons in 2009 contributed to the creation of a fully-fledged oil and gas industry. With a capacity of 10 million tons/year, the Nghi Son Refinery and Petrochemical Complex once completed will continue to write new pages in the history of development of the Vietnam's oil and gas industry.

With a total investment of over 9 billion USD, to build a complex within 4 years and ensure that it will operate safely with high quality and efficiency, it is certain that one can not expect a miracle but hundreds of cadres of the investment owners and tens of thousands of engineering staff of the EPC contractors will need to work hard: from design to manufacturing facilities around the world, and execution of construction and installation works on site with hundreds of thousand tons of supplies and equipment, as well as hundreds of million work hours to build the project.

The leaders of Petrovietnam called upon the EPC contractors with their extensive experience and solid reputation in the global oil and gas industry to implement this project as an exemplary project in terms of safety, quality and schedule. Petrovietnam will commit its best efforts, fulfil its responsibilities and obligations, and cooperate fully with the foreign partners to successfully build the Nghi Son Refinery and Petrochemical Complex into the model of modern industrial zones of the South Thanh Hoa - North Nghe An region in particular and the whole country in general.

Petrovietnam and Gazprom STRENGTHEN LONG-TERM PARTNERSHIP

On 4 October 2013, in Hanoi, Petrovietnam's leaders had a working meeting with Gazprom's Chairman of the Management Committee Alexey Miller and his delegation. At the meeting, Chairman of Phung Petrovietnam Dr.Sc. Dinh Thuc and Petrovietnam's President and CEO Dr. Do Van Hau announced the results of Petrovietnam's production in recent years, especially its efforts in the implementation of signed commitments.



Leaders of Gazprom and Petrovietnam signed an agreement on the basic principles for establishment of a joint venture company in the field of fuel gas for motor vehicles in Vietnam. Photo: CTV

At the same time, Petrovietnam and Gazprom exchanged and reviewed the results of cooperation in recent years and put forward measures to accelerate the implementation of the signed projects. In addition to further promoting co-operation in oil and gas exploration and production, Petrovietnam's leaders highly appreciated Gazprom's proposals to expand cooperation to the fields of oil refinery, supply of LNG, use of natural gas for motor vehicles in Vietnam, scientific research, training and human resource development. The parties reached the agreement to speed up the negotiation process for liquefied natural gas supply to Vietnam under the Vladivostok-LNG project and for signing a Framework Agreement on LNG supply before the end of 2013.

According to Mr. Alexey Miller, with the strong development of Vietnam's energy sector, Gazprom will invest in projects in Vietnam and Petrovietnam, and wish to promote Gazprom's activities in the country. The Gazprom Chairman said the implementation of such large-scale joint projects will strengthen the strategic co-operation between Petrovietnam and Gazprom. "I am sure it will make a significant contribution to developing the fuel and energy sector of Vietnam and providing its consumers with an environmentally friendly fuel over the long term", Mr. Alexey Miller said.

At the meeting, the two sides agreed to push up negotiations on the Head of Agreement (HOA) and an intergovernmental agreement (IGA) allowing Gazprom Neft (Gazprom's subsidiary) to involve in the project to expand and increase Dung Quat Refinery's capacity. On this occasion, Gazprom's Chairman Alexey Miller and Petrovietnam's President and CEO Dr. Do Van Hau signed an agreement on the basic principles for establishment of a joint venture company between Petrovietnam and Gazprom in the field of fuel gas for motor vehicles in Vietnam, especially for public transport vehicles in Ho Chi Minh City.

Leaders of Petrovietnam and Gazprom agreed to actively implement and expand operations in Vietnam and Russia. Gazprom will continue to implement the next exploration programme in the continental shelf of Vietnam, and pledged to actively promote the expansion of the joint venture between Petrovietnam and Gazprom (Gazpromviet) in Russia.

Nguyen Hoang

PETROVIETNAM'S OIL AND GAS PRODUCTION IN 2013 estimated at 26.18 million tons of oil equivalent

On 8 October 2013, the Vietnam National Oil and Gas Group (Petrovietnam) held a press conference to announce its production and business results of the first 9 months of 2013 and the plan for the fourth quarter. In its core areas, Petrovietnam has increased reserves by 21.81 million tons of oil equivalent (TOE) and produced a total of 19.8 million TOE, a 2.4% rise over the same period of last year. It is estimated that for the whole 2013, Petrovietnam will produce around 26.18 million TOE (16.48 million tons of oil and 9.7 billion m³ of gas).



Dr.Sc. Phung Dinh Thuc, Chairman of Petrovietnam, replied to journalists' questions. Photo: PVN

A ccording to Petrovietnam's Vice President Le Minh Hong, Petrovietnam has overfulfilled all its goals for the first 9 months of 2013, with the main production targets posting high increase compared with the same period of 2012. In the reviewed period, Petrovietnam has made 5 new petroleum discoveries, increasing oil and gas reserves by 21.81 million TOE. Total oil and gas production reached 19.8 million tons, which is equal to 103.6% of the 9-month plan, and 78.6% of the yearly plan. Of this figure, oil accounted for 12.38 million tons gas accounted for 7.42 billion m³. Besides, Petrovietnam has contributed 12.40 billion KWh of electricity to the national grid so far this year, produced and provided 1.17 million tons of urea fertiliser and 4.93 million tons of various petroleum products.

By the end of September 2013, Petrovietnam has generated a total revenue of 548.3 trillion VND and contributed to the State Budget 128 trillion VND. The value of Petrovietnam's contribution to the State budget exceeds its 9-month target by 19.3% - equivalent to 20.7 trillion VND (1 billion USD), which is very important in the context where the collection of State Budget revenues continues to meet with numerous difficulties.

Speaking at the press conference, Dr.Sc. Phung Dinh Thuc, Chairman of Petrovietnam, said that Petrovietnam will continue to strictly implement the Prime Minister's Decision No. 46/QD-TTg approving the restructuring plan for Petrovietnam during the 2012 - 2015 period as well as urge and supervise its subsidiaries to implement the restructuring plan for the 2012 - 2015 period. "Petrovietnam is now focusing on the following five key areas of operation: oil and gas exploration and production, refinery and petrochemical, gas industry, power generation, and highquality petroleum technical services. All units under the group are being rearranged in accordance with this orientation", Dr.Sc. Phung Dinh Thuc confirmed.

In the fourth quarter of 2013, Petrovietnam will continue to strictly control the progress of exploration, field development and production projects both at home and abroad, ensuring the fulfillment of plans to increase oil and gas reserves and production in the last months of 2013. Petrovietnam will put 3 new fields into production, namely Su Tu Vang Dong Bac - 5X area (Block 15-1), BK - 17 and early production in Block 67 in Peru. Besides, Petrovietnam will supervise and closely monitor to ensure the safe operation of gas transportation systems, power plants, fertiliser plants, Dung Quat Refinery, Dinh Vu Polyester Fibre Plant, and bio-fuel plants....

Under the plan, Petrovietnam will produce a further 6.38 million TOE in the last quarter of 2013, bringing the total oil and gas production of the whole year to 26.18 million TOE (16.48 million tons of crude oil and 9.7 billion m³ of gas). It is estimated that for the whole year of 2013, Petrovietnam will produce and supply 15.9 billion kWh of electricity to the national grid, 1.56 million tons of urea, and 6,279 million tons of petroleum products. Petrovietnam is expected to generate a total revenue of 732 trillion VND and contribute 170 trillion VND to the State Budget.

Ngoc Anh

PALEOGENE SEDIMENT COMPLEX - GEOLOGICAL FACTORS IMPACTING ON ITS RESERVOIR QUALITY AND HYDROCARBON POTENTIAL

Ngo Thuong San¹, Cu Minh Hoang², Phung Khac Hoan² ¹Vietnam Petroleum Association ²PVEP Overseas

Summary

The Paleogene reservoir complex has diverse facies which change rapidly both laterally and vertically. Its depositional environments are determined as alluvial, deltaic, tidally affected lacustrine and, rarely, with a shoreline. The variety in sedimentary facies and lithological heterogeneity are the main causes for the anisotropy in porosity and permeability of the Paleogene reservoirs, thus reducing field exploration and development efficiency as well as the application of secondary exploitation in order to increase the oil recovery.

Reservoir quality has resulted from the combined effects of primary diagenetic elements and secondary alteration. Secondary deformation has a bidirectional influence on the reservoir quality, reducing the primary porosity but also forming secondary porosity. Dual porosity is one of the important properties of the Paleogene reservoir that should be considered in production management.

Secondary fractures could potentially increase the flow in the tight Paleogene reservoirs.

The production from Paleogene reservoirs is still small, not corresponding to the potential of Paleogene formations as rich source rocks. The Late Eocene-Oligocene reservoir, therefore, needs to be investigated and sufficiently evaluated as a potential hydrocarbon bearing play in order to enhance production and reserves.

1. The lithology of the Paleogene reservoir complex and its heterogeneous character

In the Cuu Long basin, wells are concentrated on structural uplifts, thus encountering only the upper strata

of Paleogene age, mainly those of of Late Eocene-Oligocene age.

The Late Eocene-Oligocene assemblage was found in many locations with a full or partial section and showing high facies heterogeneity (Fig.1):

- The underlying Tra Cu formation of Late Eocene - Early Oligocene age, corresponding to the seismic sequence E, is composed mainly of fine- to coarse-grained sandstones interbedded with siltstone, shale, and thin layers of coal and intraformational conglomerate; - The overlying Tra Tan formation of Late Oligocene age, equivalent to seismic sequences D and C, is characterised by finer-grained sedimentary rocks, mostly shale and siltstone intercalated with thin layers and lenses of sandstone.



Fig.1. Stratigraphic column of Tertiary formation - Cuu Long basin

The Paleogene sandstones are hydrocarbon reservoir with low flow rate due to low permeability. However, some wells yield high flow, at times reaching 2 - 3 thousand barrels per day, such as at North Bach Ho, and Southeast Rong...

1.1. The sandstone of Sequence E, (Fig.2) grey and light brown colored with cross-bedding, blocky shape, and the grain size of 0.25 - 1mm, is overlain by fine- to coarse-grained sandstones intercalated with siltstone, coal bearing argillite, and shaly schist with thin coal layers. Sandstones are polymictic with compositions varying from arkose, lithic arkose to litharenite, feldspar litharenite, and with increasing tendency for the presence



Fig.2. Fine to medium-grained sandstone oil-saturated moderately sorted with cross-bedding point bar facies - Early Oligocene - BH-10 well - 4,054m

of rock fragments to the Northeast of the basin (Block 01-02, 15), where they were formed plausibly in a terrestrial environment of high energy, close to the supply source. The facies changes rapidly from proluvium, channel bar to alluvial plain and swamp facies laterally, towards the depositional centre, and stratigraphically upwards.

| | | reserven re | | | ciu | | | | | | | |
|----------------|---------------|-------------|--------|----------|-------------------|---------|--|--|--|--|--|--|
| | Clay (%) | | | | | | | | | | | |
| Unit | Smectite | Kaolinite | Illite | Chlorite | Mixed minerals | Zeolite | | | | | | |
| Early Miocene | | | | | | | | | | | | |
| 23 | 36 | 40 | 11 | 8 | 5 | - | | | | | | |
| 24 | 19 | 66 | 6 | 8 | 1 | - | | | | | | |
| Late Oligocene | | | | | | | | | | | | |
| I | - | 13 | 39 | 36 | 12 | - | | | | | | |
| П | - | 5 | 35 | 43 | 15 | 2 | | | | | | |
| | Early Miocene | | | | | | | | | | | |
| VI | - | 4 | 39 | 40 | 14 | 3 | | | | | | |
| VII | - | - | 20 | 53 | 27 | - | | | | | | |
| VIII | - | - | 28 | 55 | 12 | 5 | | | | | | |
| IX | - | 2 | 30 | 52 | 6 | 10 | | | | | | |
| Х | - | - | 28 | 58 | 3 | 10 | | | | | | |
| XI | - | - | 10 | 80 | - | 10 | | | | | | |

Tab.1. Distribution of clays minerals in Miocene - Oligocene reservoir rocks in Bach Ho field



Fig.3. Interfingering character of stratigraphic arrangement in the Early Oligocene section - Bach Ho field



Fig.4. Porosity - Permeability relationship of Oligocene reservoir in Cuu Long basin

The high concentration of feldspar (plagioclase) and the presence of unstable, ductile minerals in the cement composition, such as albite, zeolite, illite, chlorite, and hydromica, as well as compaction and secondary alteration had reduced the size and volume of primary porosity. An abrupt increase in the zeolite and decrease in kaolinite contents has been recorded in the 3,700 - 4,400m interval (SH VI-IX) (Tab.1). This phenomenon is observed in a number of wells in the Bach Ho, Rong and Rang Dong structures.

Facies-lithological studies indicate that the E sequence consists of fluvial, partly lagoon-deltaic clastic sediments deposited in sub-grabens. Dominant facies are fluvial bars of braided and meandered rivers, alluvial fans and coastal sediments bordering sparsely distributed lagoons.

Alluvial fans were deposited under the influence of strong currents, which flowed in multi-direction streams. Shale laminae between sand lenses are confining and do not play the role of a seal. The evidence of desiccation cracks and bioturbation by burrowing organisms shows that the water was shallow and occasionally receded or evaporated.

Lithologic heterogeneity due to facies variation has resulted in the interfingering character of stratigraphic arrangement in the Early Oligocene section (Fig.3).

The average porosity of the Early Oligocene sandstone is 12 - 14%, and 18% in some places; while permeability ranges from 1 - 50mD. Permeability tends to be better in the upper part of the section and decreases rapidly with depth, while porosity is normally below 10 - 12% at 4,000m, where strong quartz overgrowth and chloritisation occurred... The quality of that sandstone reservoir is normally poor with low flow rate, but sometimes the flow could reach several hundreds of tons of oil per day as in Bach Ho and Southeast Rong, or condensate gas in Emerald and Su Tu Trang fields (Fig.4).

1.2. The rocks comprising sequence D in the Late Oligocene Tra Tan formation are mainly bituminous shale, coaly shale, lignite and siltstone with intercalated sandstone lenses of various thicknesses, in which clastic components include quartz, feldspar and rock fragments. The amount of orthoclase outweighs that of plagioclase - that is a significant property compared to sequence E's sandstone. The sandstone belongs to the arkose and lithic arkose clan, medium to coarse-grained, medium sorted and of poor roundness. Authigenic minerals include guartz and calcite, whilst secondary clay minerals are illite and chlorite. The content of zeolite increases with a decrease in the composition of kaolinite. A high variety of rock texture was observed such as fine crossbedding, planar bedding and massive. The depositional environment is determined to be a meandering river and low deltaic plain environment of medium energy. The sequence D is considered as the regional source and seal of the Oligocene assemblage although it contains a certain amount of sandstone layers, the latter are of small in quantity and are normally thin and confining. On the basin boundary, where shale facies is changed into sand facies, the sandstone layers, especially in the lower part of sequence D, become a target of interest.

1.3. Sequence C in the Late Oligocene Tra Tan formation is commonly composed of shale, coaly shale rich in organic matter, thin coal layers, and siltstone interbedded with thin sandstone lenses. Yet, in several structures such as Diamond, Ruby, Su Tu, Rang Dong, and Bach Ho fields, flow of high charge is found in areas where the number and thickness of these sandstone layers increase. Sandstones are polymictic and belong to the arkose and lithic arkose type with a content of quartz (40-62%), feldspar (30-50%), and rock fragments (9 - 13%). Particularly, in the Su Tu area, the litharenite and feldspar litharenite are dominant with the rapid increase of rock fragments ratio (40 - 50%) while the content of feldspar decreases to 5 - 15%. Sandstones are medium- to coarse-grained with medium sorting and good roundness. Cement is composed of authigenic quartz and secondary clay minerals such as chlorite and illite. The amount of illite is recorded as increasing rapidly and becoming dominant below 3,300m with a decrease in kaolinite content. The rocks are cross bedded and rippled. Porosity ranges between 12 - 21% with an average of 14%. Permeability is 2 - 26mD, and reaches 50 - 60mD in certain places. The depositional environments were swamps, floodplains, and anoxic tidal plains affected by sporadic sea ingression from the Northeast.

The facies variety and lithological heterogeneity are the main causes for the anisotropy in porosity and permeability of the Paleogene reservoir, reducing field exploration and development efficiency as well as limiting the application of EOR methods.

2. Geological factors impacting on the reservoir quality of Paleogene formation

Many factors of different origins could affect the reservoir quality. However, reservoir quality is a combination of the effects of primary elements and secondary deformation during diagenesis. The quality of Paleogene reservoirs is generally low.

2.1. Sedimentary lithology and reservoir characteristics

Such primary elements related to the sedimentary lithology as rock texture (grain size, sorting, and roundness), rock type (clastic and cement, clay minerals), sand-shale ratio, reservoir thickness and distribution significantly affect the porosity and permeability of the Paleogene reservoir complex, especially that of the Eocene - Early Oligocene sequence E.

- There is a relationship between porosity and permeability, which tends to decrease with depth due to loading compaction. The visual porosity is normally less



Fig.5. Functional relation between porosity versus depth of Miocene/Oligocene reservoir rocks

than 18%. The effective porosity of sandstone is usually less than 10% with permeability less than 10mD. The granular porosity is less than 4% below 4,350m in Bach Ho field (Fig.5).

- The values of porosity and permeability are functions of grain size, sorting, abrasion, clastic components and textural arrangement. The porosity of medium- and coarse-grained sandstone is generally low because the clasts are poorly sorted and poorly eroded; the development of clay minerals in the cement composition such as illite/chlorite and zeolite indicates the level of secondary alteration and the ability to fill up primary



Fig.6. Non-uniform character of oil contamination of Oligocene reservoir - Bach Ho field



Fig.7. Distribution of overpressure zones with depth in Cuu Long basin

pores, thus reducing the Oligocene reservoirs' porosity and permeability.

- In terms of clastic composition, the Oligocene sandstones are mostly polymictic. They are classified as arkosic and lithic arkosic sandstones with high contents of unstable fragments, such as feldspar (20 - 40%) and particularly fragments of acidic extrusive rocks, granitoid and shaly schists (7 - 25%). Such high concentration of unstable rock fragments is one of the factors that have reduced the reservoir quality of this terrigeneous sediment.

- Another critical factor is the cross-bedding of the reservoir rocks. Analysing the cross-bedded reservoir rocks for intergranular porosity, the vertical variation is firstly investigated, while laterally the reservoir is considered homogeneous at least in the extended space between correlated wells. The discrepancy in reservoir quality is complicated, especially in cross-bedded strata. Cross-bedding has a direct influence on the porositypermeability (K-q) relationship. At the same porosity, a reservoir rock has maximum permeability and thus yields the maximum flow in the direction parallel to that of the crossbedding, while in the perpendicular direction, the permeability and flow have their minimum values. The difference is recorded to be tens of times. Fig.6 shows that the oil contamination is not uniform but depends on sorting, grain size and texture of the reservoir rocks.

- Abnormal overpressure is another factor that influences the distribution of reservoir porosity and permeability. Abnormal overpressure greatly affects the petroleum system, including trap seal integrity, reservoir quality, timing of maturation of the source rock, as well as the timing and the direction of hydrocarbons' primary migration (Fig.7).

Reservoir quality is closely related to the ability of clastic rocks to lose porosity under mechanical loading compaction, which increases with depth. However, abnormal overpressure could reduce, and at times stop the mechanical compaction, preserving the porosity in sandstone reservoirs interbedded with or underlying overpressured shale. Abnormal overpressure could also retard diagenesis. Abnormal overpressure was commonly present in the center of the basin where thick shale was deposited. In the proximal part of basin the grain size becomes coarser, the thickness of shale decreases, and the overpressure phenomenon disappears. Abnormal overpressure is always a favorable condition for both lateral and upward vertical migration of hydrocarbon within reservoir rocks while interbedded with overpressured shale, but the pressure differentiation could also create migration to underlying reservoir sequences.

2.2. Bidirectional influence of the secondary deformation on the reservoir quality

Paleogene, especially Eocene-Early Oligocene, sediments were deeply buried and are distributed below 3.5-4.5km. Under the high pressure of loading compaction and high temperature, the Paleogene sediments have undergone vigorous secondary deformation, changing to the late catagenesis - early metagenesis stage, which is manifested in two opposite processes that result in a reduction of the primary porosity and the formation of secondary porosity.



Fig.8. Pore spaces infilled by clay minerals, but still with good connectivity (ST-3X well)



Fig.9. Early Oligocene sandstone highly fractured - Bach Ho field fractures filled by secondary minerals

- The cementation process, especially quartz cementation and the formation of secondary minerals to content of more than 20%, sometimes 25 - 30%, causes the infilling of primary pores and reduction of the intergranular porosity (Fig.8). According to the field statistics from Blocks 01-02, 09, 15, and 16, with



Fig. 10. Core at 2,993.25m in E sequence of Rang Dong field



Fig.11. Relation between P_c and S_w of the Late Oligocene reservoir - Bach Ho field



Fig.12. Relation between P_c and S_w of the Early Oligocene reservoir (E sq.- ST field)

the content of secondary minerals more than 30%, the primary reservoir porosity should be reduced to less than 5%, equivalent to the level of deformation from late catagenesis to early metagenesis, which occurs at the 4,200m depth. At this depth, oil has low mobility and usually loses its flow ability, particularly in the case of medium - heavy oil (29 - 35°API) with low GOR (300 - 350scf/stb).

- Contrary to the above trend, secondary deformation can also generate secondary pore spaces, an important factor that helps increase the porosity and permeability of Paleogene clastic sediments. The appearance of microfractures and micropores during late diagenesis, especially when changing from late catagenesis to early metagenesis due to dissolution, chemical solution on the pore walls, and volumetric contraction due to the formation of secondary minerals, had initiated the generation of secondary porosity and permeability. The lithological analysis reveals the existence of vuggy porosity with

> radius of 0.2 - 0.35mm; and microfractures with apertures up to 1mm mostly filled with secondary minerals (Figs.9 and 10). For reservoir rocks of early catagenesis the intergranular porosity plays a critical role, while for clastic sediments in the late catagenesis - early metagenesis stage, secondary pores (vuggy pores, fractures) become the potential hydrocarbon migration pathways.

> - However, besides the ability of late diagenesis to form secondary deformation which increases the reservoir quality, differential fracturing of brittle clastic rocks in the intercalated sand/shale reservoir sequence, formed during the folding and faulting period of Late Oligocene tectonic compaction and inversion, could also form secondary porosity and permeability in both structural and stratigraphic traps.

Secondary fractures could potentially increase the flow of the tight Paleogene reservoirs

The influence of fractures, which are always present in clastic rocks, in improving low fluid flow rates has not yet been as sufficiently investigated as in fractured basement reservoirs. The impact of the presence of fracturest is also demonstrated by the abrupt change on the capillary curves of reservoir rocks with different values of permeability (Figs.11, 12 and 13).



Fig. 13. Relation between P, and S, - Early Oligocene reservoir Bach Ho field



- Across Hinge In Dip Directio
- As With "A" But in Forelimb
- Alternate to "B" Oblique To Strike & Dip Parallel To Strike in Backlimb In Most Fractured Layer (s)
- D Oblique To Both Secondary Hinge and Layering

Fig.14. Fracture distribution on a folded anticline trap [4]



Fig. 15. Map illustrating the distribution of total porosity in Early Oligocene sequence of Bach Ho field (data VSP)

Thus, the characteristics of fracture distribution need to be analysed in order to identify at what location and when the fracture system would dynamically increase the flow. In the case of fractured basement, the density and connectivity of the fractures system network depend upon the stress field's attributes and its intensity while in the stratified clastic reservoir, the fracture intensity, spacing and fracture width depend upon: (1) lithology, (2) grain sizes, (3) rock texture, (4) porosity, (5) reservoir thickness and (6) the location of fractures on the folded structures.

The study results indicate that:

- Rocks with a high content of ductile minerals have denser fracture distribution, but narrower fracture width as comparing to those of brittle composition. This could explain the fact that high flow reservoirs in the Cuu Long basin are normally quartz arkose sandstones, quartz polymictic, litharenite of high brittle clastic and low feldspar ductile contents. Fracture width and connectivity become better.

- In general, there is a relationship between a decrease in grain size and an increase in fracture density, probably due to the fact that fine-grained layers are more thinly-bedded as compared to the thickly-bedded and blocky coarse-grained layers.

- Rocks of the same composition and texture, but of high tightness and low porosity, will have sparse fracture networks, but open fractures possibly become wider.

- With respect to all lithological and sedimentary characteristics, given the same compaction condition, thin sand layers tend to be more fractured with closer spacing.

- The fracture intensity depends upon the stress field formed during the folding and faulting tectonic deformation. The role of faulting in stratified traps is investigated for sealing capacity rather than fracture extent. Moreover, with the multi-staged property of the fault system in the Cuu Long basin, the closure of the Late Oligocene seal, containing thin shale strata, is still a big risk. Therefore, the possibility for the fracture system to act as a factor to increase the porosity and permeability of the Paleogene reservoir is only related to the folding movement, and in the case where the covering Late Oligocene shale as a regional seal was not destroyed. The fracture system is usually very complicated and is extrapolated from surface field studies. The variation in fractures' location and intensity depends upon the shape and origin of the folds. Their trend and plunge could either be parallel or perpendicular to the bedding surface. Despite their diversity and complexity, fractures of high dip tend to be concentrated in the apex and along the axis of the folds, and sparsely extended on the two limbs. The strike of fractures is usually parallel to the fold axis (Fig.14).

This trend is very critical to locating the exploration and production wells in clastic traps such as those of Paleogene age.

- Also, the abrupt change on the relation curve between capillary pressure P_c and saturation S_w possibly indicates the presence of the dual porosity character of the Paleogene reservoir: the fracture porosity as open conduit of high permeability and the matrix intergranular porosity having lower permeability.

Though the relationship between fracture distribution and the porosity character of the Paleogene reservoir of Bach Ho field has not been fully investigated, some correlation can be observed between the porosity distribution and the location of the apex zone of the anticlinal axis, and also partially with faults (Fig.15).

2.3. Hydrocarbon potential distribution in the Paleogene reservoir formation

Commercial oil production from the Oligocene rocks is identified systematically at Bach Ho field of Cuu Long basin. The oil potential Oligocene formation is developed mainly in the Northern part of Bach Ho field and overlapped the Mesozoic intrusive granite basement. The oil trap structure is complicated, of multilayered character and isolated mostly by faults and lithological barriers (Fig.16).

In general, the Oligocene reservoir complex could be grouped into Late and Early Oligocene productive formations with different reservoir properties. Up to now the Early Oligocene reservoir is the main production target for Vietsovpetro.



Fig. 16. Multi-layered character of Early Oligocene reservoir - Bach Ho field (VSP data)



Fig.17. Distribution of hydrocarbon potential with depth (data VSP)

Some common trends in reservoir quality can be recorded:

 Oil bodies are discontinuous and heterogeneous in reservoir quality. They are separated by faults, and sealed by tectonic and lithological barriers.

- The highest block is less compacted and the western part, more impacted by faults is better qualified in reservoir quality.

- High heterogeneity of poro-permeability with both area and depth and poor hydrodynamic connectivity between wells, negatively affected the result of waterflooding.

- Exploitation is performed mainly by the displacement mechanism of water drive with local gas solution drive.

- High productivity seems to be related to the apex zone of the block anticline and partially to the fractured zone close to faults where the reservoir quality is plausibly better.

- In most of the wells, the production index varied weakly, but tends to decrease with time.

- For exploitation, high interbedding and reservoir heterogeneity, negatively affect the reliability of reservoir petrophysic parameters and consequently the assessment of reserves and oil distribution in each block and every productive unit, and also the design of offshore facilities.

- Averaging the production in Oligocene producer wells by depth intervals (data to 2006), the output capacity of the Oligocene reservoir is estimated (Fig.17):

- Reserves recovered at depth 3,450 - 3,500m are the highest with the oil output of about 1,056 thousands tons, taking 20% of total oil production from Oligocene reservoir.

- The most effective producing interval is at depth 3,300 - 4,250m.

- Production capacity decreases with depth.

3. Conclusions

- The lacustrine-delta facies complex which developed in rift basins during the Paleogene period, particularly in the Cuu Long basin, has a great hydrocarbon potential. - The Paleogene reservoir rocks have diverse facies, which varied rapidly in both horizontal and vertical directions, but those of high production are underlying the sequence D's shale (seal) or thinly interbedded in the D and C sequences. They were formed in the environment of alluvial plain, lacustrine delta or coastal plains regularly affected by tidal influx.

- The variety in sedimentary facies and lithological heterogeneity, the great variation of net to gross sand ratios and interbedding coefficient in lateral and vertical direction are the main causes of the anisotropy in porosity and permeability of the Paleogene reservoirs, reducing field exploration and development efficiency as well as the application of secondary exploitation in order to increase the oil recovery.

- Being deeply buried and having undergone the effects of loading compaction and high temperature, the Paleogene reservoir rocks, particularly of the F and E sequences, are usually tight, cemented, and subject to a high level of diagenesis and secondary deformation (from late catagenesis to early metagenesis). The reservoir quality of these strata is usually low.

 The values of porosity and permeability depend upon grain size, sorting, roundness and clastic composition.

- The porosity and permeability quality tends to increase in the overlying part of the cross section and rapidly decrease with depth. Due to strong zeolitisation and chloritisation, porosity is normally less than 5% at 4,200m and below.

- Another critical factor is the cross-bedding of the reservoir rocks. Cross-bedding has direct influence on the porosity-permeability (K- ϕ) relationship. At the same porosity, reservoir rock has the maximum permeability and thus yields maximum flow in the direction parallel to that of the crossbeds, while in the perpendicular direction, the permeability and flow are minimum at a minimum.

- The abnormal overpressure developed in the shale of the Paleogene sequence D affects the distribution of porosity and permeability. Abnormal overpressure always creates favourable conditions for lateral and vertical fluid migration in the reservoir sequence.

- Fractures are always present in the clastic rocks. The differential fracturing of brittle clastic rocks formed in the reservoir formation comprising interbedded shale and sandstones during the folding-faulting process in the Late Oligocene tectonic compression and inversion could generate secondary porosity and permeability in both structural and stratigraphic traps.

- The density and spacing of secondary fractures depend upon: lithology, grain size, porosity, reservoir thickness, and location of the fracture system on folded structures.

- Secondary fractures are a potential factor that could increase the flow in tight Paleogene reservoirs. The intensity of fractures with high dip tends to be concentrated on the structures crest and along the axis of the folds, and decreases on the two fold limbs. The strike of fractures is usually parallel to the fold axis. This trend is very critical to locating the exploration and production wells in clastic reservoirs.

- Dual porosity is one of the important properties of the Paleogene reservoir that should be considered in production management.

- The high flow rate in the Oligocene-Late Eocene reservoir is possibly related to: firstly, the secondary fractures, that were associated with inversion folding at Late Oligocene; secondly, the low viscosity light oil, or gas and condensate. The oil viscosity and density plausibly affect the flowing capability of high rate. - Up to now about 300 million tons of oil have been produced in Vietnam, but the production percentage from Paleogene reservoirs still does not exceed 7%, and does not correspond to the potential of Paleogene formation as source rock.

- The Late Eocene-Oligocene reservoir needs to be investigated and sufficiently evaluated as a potential hydrocarbon-bearing play in order to enhance the production and hydrocarbon reserves of Vietnam.

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CHARACTERISATION OF CRUDE OILS AND GAS IN BLOCKS 05-2 AND 05-3 - NAM CON SON BASIN USING BIOMARKERS, GAS CHROMA-TOGRAPHY AND CARBON ISOTOPE ANALYSIS

> Nguyen Thi Bich Ha, Ha Thu Huong, Nguyen Thi Thanh Vietnam Petroleum Institute

Summary

Research on petroleum systems is the basis for evaluating the potential of a sedimentary basin during exploration and exploitation processes. In this, results from analysed samples are very useful. In the Nam Con Son basin offshore Vietnam, several oil and gas fields have been found such as Dai Hung, Thanh Long, Bo Cau, Nguyet Thach, Moc Tinh, Hai Thach, Lan Tay, Lan Do, etc. However, the origin of organic matter and the distribution of crude oils in Nam Con Son basin still pose difficult problems for petroleum geologists. Certain questions remain to be answered, such as the origin and transformation level of organic matter, depositional conditions, rate of sedimentation or even why geologists have not discovered oil accumulations leading to gas being found more than oil. In this paper, the authors use advanced analysis technology and combine effective analysis parameters to evaluate and classify the origin of oil and gas in Blocks 05-2 and 05-3. According to the results of GC, GCMS and carbon-13 isotope analysis, crude oil at the Nguyet Thach well is believed to be generated from a low maturity source rock containing lacustrine organic matter.

Evaluating and classifying oil groups based on the results of physical characteristics, chemical composition analysis and more detail analysis such as GC, GCMS of crude oils and condensates can indicate the depositional and degrading environments of organic matter as well as the maturity level of source rock which generated these oils and condensates. Specially, the results of carbon isotope analysis of crude oil and gas samples clearly prove the origin of oil and gas as well as gas-oil and oil-oil correlation. In Blocks 05-2 and 05-3, oils have been found in Late Miocene, Middle Miocene and Early Miocene aged reservoirs at wells 05-2-HT-2X, 05-3-MT-1X, 05-2-NT-1X, 05-2-B-1X, 05-2-KCT-1X, 06-LD-1X, and 06-LT-1X OBS1.

1. The primary physical and chemical characteristics of oil and condensate

The physical characteristics and chemical composition analyses of oil and condensate samples in Blocks 05-2 and 05-3 are listed in Table 1.

Of the 21 samples taken from Block 05, there are 8 oil samples and 13 condensate samples which have API more than 45 (Table 1). Both DST#3 oil sample (2,874 - 3,004m) in Block 05-1a and RFT5 sample (2,864.5m) are heavy oils (°API < 25), 6 samples are normal oils with API varying from 25 -45. Almost all oil samples in the Nam Con Son basin have low sulfur, vanadium and nickel contents. This is typical for oils derived from mainly higher plant organic matter.

The relationship between sulfur content and API shows that oil samples belong to B, C, D and E types. In addition, the Pr/Phy ratios also illustrate type D and E of these samples. According to the oil classification of BP Reach (Table 2), if a crude oil has an amount of saturate hydrocarbon fraction more than 70%, it will belong to D type and vice versa. In Table 1, all crude oils and condensates of Block 05 belong to type D which were generated from source rocks containing mainly higher plant organic matter.

2. Gas chromatography and gas chromatography-mass spectrometry

The results of gas chromatography and gas chromatography-mass spectrometry analysis are used as fingerprints to identify the type of organic matter in oil and rock samples. Pristane and Phytane are separated from fractions of Chlorophyll. The presence of these components reflects the increase in activity of bacteria in a primary Table 1. The primary physical and chemical characteristics of oil and condensate in Block 05, Nam Con Son basin

| iu Nickel | (mqq) (m | | ~ | $\overline{\nabla}$ | ~ | 2.45 | 2.82 | 1.84 | | 5 | 31.00 | | | | | | | | 1.30 | 2.56 | | |
|-----------|------------------|------------|-------------------------|----------------------|---------------|--------------|-------------------|------------|-----------------|-----------|---------------|------------------------------|---------------|---------------|---------------|---------------|---------------|-----------------|-------|-------------|---------|---------|
| Vanad | ıdd) m | | $\overline{\checkmark}$ | ~ | ~ | < 0.1 | < 0.1 | < 0.1 | | < 2 | < 2 | | | | | | | | 0.02 | 0.07 | 0.22 | 0.229 |
| Phy/ | nC ₁₈ | 0.24 | 0.24 | 0.16 | 0.12 | 0.17 | 0.19 | 0.19 | 0.18 | 0.33 | 0.75 | 0.20 | 0.17 | 0.14 | 0.14 | 0.17 | 0.18 | | | 0.26 | 1.36 | 1.37 |
| Pr/ | nC ₁₇ | 1.48 | 1.49 | 0.85 | 0.63 | 1.21 | 1.30 | 1.38 | 1.22 | 1.84 | | 1.11 | 0.98 | 0.91 | 0.87 | 1.07 | 0.76 | | | 1.39 | 5.92 | 6.14 |
| Pr/ | Phy | 5.33 | 5.57 | 5.52 | 5.20 | 6.82 | 6.31 | 6.93 | 6.78 | 5.43 | | 5.65 | 5.95 | 6.50 | 6.04 | 6.26 | 4.32 | 6.40 | 5.34 | 4.48 | 14.91 | 14.84 |
| NOS | (%) | 3.68 | 1.94 | 1.81 | 1.17 | | 0.10 | | 0.10 | 0.20 | 15.00 | | | | | | | 9.66 | 8.13 | 6.64 | 85.09 | 85.16 |
| HCaro | (%) | 20.23 | 25.43 | 26.17 | 20.23 | 10.50 | 17.20 | 6.90 | 10.90 | 24.10 | 40.20 | | | | | | | 8.66 | 13.82 | 19.94 | 18.92 | 20.00 |
| HCsat | (%) | 76.09 | 72.63 | 72.02 | 78.60 | 89.50 | 82.70 | 93.10 | 89.00 | 75.70 | 48.80 | | | | | | | 82.68 | 78.05 | 73.41 | 66.17 | 65.16 |
| S | (%) | | 0.086 | 0.058 | 0.046 | 0.05 | 0.05 | 0.05 | | 0.10 | 0.20 | | | | | | | 0.096 | 0.049 | 0.01 | 0.03 | 0.02 |
| IdΦ° | | | 32.86 | 38.06 | 40.50 | 36.15 | 35.90 | 36.40 | | 32.80 | 17.30 | 30.90 | 34.00 | | 39.20 | 25.20 | 30.20 | 36.18 | 31.75 | 35.10 | 38.10 | 37.44 |
| Type of | sample | Crude oil | Crude oil | Condensate | Condensate | Condensate | Condensate | Condensate | Condensate | Crude oil | Crude oil | | | | | | | Crude oil | | Crude oil | | |
| Reservoir | formation | 01212 | Miocene | Early Miocene | Oligocene | Late Miocene | Middle Miocene | | Early Miocene | | Early Miocene | Early Miocene Middle Miocene | | Miocene | | Late Miocene | | | | | | |
| Depth | (m) | 3,198.5 | 3,217.3 - 3,222.5 | 4,092.2 - 4,097.3 | 4,559 - 4,575 | 2,956.5 | 3,195.8 | 3,497.2 | 3,617 | 3,765 | 2,864.5 | 2,594 - 2,605 | 2,594 - 2,637 | 2,651 - 2,663 | 2,737 - 2,749 | 2,874 - 3,004 | 2,984 - 3,004 | 3,057 | 3,069 | 2,849.5mBRT | 3,075.5 | 3,136.5 |
| Samula | | RFT | DST#3 | DST#2 | DST#1 | MDT | MDT | MDT | MDT | MDT | RFT5 | DST#6 | DST#6A | DST#5 | DST#4 | DST#3 | DST#2 | RFT | - | RFT | DST#2 | DST#1C |
| Well name | | 05-2-HT-1X | | | | 05-2-HT-1X | 1 | | 05-2-NT- 1RX | | 1 | 05-1a-DH- | 4X | 1 | | 05-2-KCT- | 1X | 05-3-MT- 1RX | | 05-3-MI-1X | | |



Fig. 1. Relationship diagram of API versus sulfur content as well as Pr/Phy ratio of Blocks 05-2 and 05-3 and surrounding areas

| 0.1 | Overania matter innut into | Denesitien environment | Effects on oil properties | | | | | |
|-------|---|---|---------------------------|---------------------------|--|--|--|--|
| class | kerogen | of source | Organic matter input | Deposition environment | Either/Both of there | | | |
| A | Phytoplankton Bacteria | Marine (carbonate and other non-siliciclastic | Low content | High content | Most other | | | |
| В | Phytoplankton Bacteria | Marine (Siliciclastic sediment) | Low content | Moderate content | controlled by a (sometimes poorly understood) | | | |
| С | Phytoplankton Bacteria | Lacustrine (fresh water) | High wax content | Low content | | | | |
| D | Higher land plant (Angiosperm dominated) Debris, Bacteria | Non marine | High wax content | Low content | | | | |
| E | Higher land plant (gymnosperm dominated) Debris, Bacteria | Non marine | High wax content | Low content | combination of factors | | | |

Table 2. Oil classification of BP Research Centre, Sunbury [3]

oxidising environment. However, this ratio is influenced by changes in rocks containing organic matter. Moreover, in the maturity process, Phytane is generated faster than Pristane. Therefore, this ratio decreases with maturity level. Pr/Phy ratio of isoprenoid chain is used to evaluate the origin and environment of organic matter [3].

Biomarkers in Triterpane and Sterane distributions of GCMS analysis help geochemists determine that the source of organic matter is higher plant, lacustrine algal or marine algal. Furhermore, we can also know the time that source rocks generated oils and that source rocks are Tertiary aged or older. These results play an important role in research on the depositional environment, building calibration parameters for a maturity model of source rocks as well as identifying the oil-oil and oil-source rock correlation. It is a useful tool in exploration and exploitation processes.

The results of GC analysis from 21 samples show that these samples have similar organic matter origin which related to source rocks containing terrestrial organic matter, deposited and degraded under oxidising and weakly reducing conditions (Fig.2). The Oleanane and Ts/Tm ratio diagram (Fig.3) was used to distinguish lacustrine algal and fluvio-deltaic source of organic matter. In this figure, almost all crude oils and condensates found in block 05 and surrounding areas locate at the estuarine-fluvio deltaic zone and mix of lacustrine and fluvio-deltaic zone, with only a few samples in the lacustrine zone.



Fig.2. Pr/nC_{17} and Phy/nC_{18} ratio diagram



Fig.3. Oleanane and Ts/Tm ratio diagram



in Moc Tinh structure [2]

In the Moc Tinh structure, crude oils were found at the MT-1X and MT-1RX wells. According to the results of GC analysis for the saturated hydrocarbon fraction of 2 crude oil samples DST#1c and DST#2 at the 05-3-MT-1X well and RFT#1c sample at 2,849.5mBRT at the 05-3-MT-1XR well, all three samples have C15+ distribution of saturated hydrocarbon in arch shape. This is typical for mature organic matter (Fig.4a). In addition, all these samples have high Pr/Phy ratios (6.14; 5.92 and 4.48 respectively). Therefore, the source rocks have been mature and contained mainly higher plant organic matter (with kerogen type 3).

In Triterpane distribution in m/z 191 fraction, Moretane and Oleanane constituents are typical for organic matter derived from higher plants. All samples have Oleanane (peak B) and Bicadinane constituents which are reliable signals of terrestrial organic input. Bicadinane is related to plants containing resinite which was found in oils of Southeast Asia. Oleanane which is related to Angiosperms was found only in Cretaceous or younger aged oil and rock samples (Figs.4b and 4c).

In Sterane (m/z 217) distribution, the domination of C_{29} with C_{27} and C_{28} shows the abundance of terrestrial organic matter. Ts and Tm constituents (Figs.4b and 4c) are parameters for evaluating the maturity. In fact, when the maturity increases, the Tm transforms into Ts. Both oil samples have an amount of Ts higher than that of Tm, that means these oils were expelled from highly mature source rocks.

In the Kim Cuong Tay structure, two RFT crude oil samples were found at 3,057m and 3,059m in Middle Miocene reservoirs. Both of them have high Pr/Phy ratios (6.4 and 5.34 respectively).

The Sterane (m/z 217) and Triterpane (m/z 191) distributions of these samples are shown in Figs.5a (RFT 3,057m) and 5b (RFT 3,059m). These distributions have a large amount of Bicadinanes and Oleananes. The Ts/Tm ratios suggest that both of them were generated from source rocks in the oil window phase. The Hopane distribution (Fig.5b) is different from extended Hopane distribution in which the height of C_{31} to C_{35} peaks decrease with the increase of the number of carbon atoms. It also illustrates that these source rocks were deposited and degraded in a stronger reducing environment.

Four condensate samples and one oil sample were found in the Hai Thach structure. The results of saturated hydrocarbon GC analysis of these samples show that they have abundant n-alkane fraction and rather high



Fig.4b. The results of GC and GCMS analysis of crude oil sample (DST#2 - 3,075.5m) in Miocene reservoir at the 05-3-MT-1X well [2]



Fig.4c. The results of GC and GCMS analysis of crude oil sample (DST#1C, 3,136.5m) in Miocene reservoir at the 05-3-MT-1X well [2]



Fig.5a. The results of GCMS analysis of crude oil sample (RFT, 3,057m) in Middle Miocene reservoir at the 05-2-KCT-1X well [2]

Pr/Phy ratios (from 5.43 to 6.93). Therefore, their source rocks contained mainly organic matter derived from higher plants (Table 1).

They also have similar Sterane (m/z 217) and Triterpane (m/z 191) distributions which have abundant Bicadinanes and Oleananes. Very clearly seen are the very high Oleanane/C30 ratios of all these samples (from 114.68 - 130.79). In addition, the Ts/(Ts + Tm) ratios, also rather high (0.4 - 0.47), show that these samples were derived from highly mature source rocks.

The differences between condensates and oil in the Hai Thach structure and oils/condensates in the Moc Tinh, Kim Cuong Tay and Nguyet Thach structures are the majority of C_{29} in Triterpane (m/z191) distribution and the presence of C_{25} and C_{30} Norhopane. These are biosignals of marine organic input.

The results of whole oil GC analysis of the RFT oil sample at 2,864m in 05-2-NT-1X clearly show that the light hydrocarbon fraction was lost. The Pr/Phy ratios from the saturated hydrocarbon GC analysis are low. This is different from those of oils above.

The presence of C_{30} 4 methyl Steranes and Diasterane constituents is typical of the Stearane distribution which shows the trace of lacustrine organic matter and rich shale environment. In Triterpane distribution, Bicadinane constituent is quite rich, in contrast, Oleanane is absent. In general, oils could be generated from lacustrine source rocks containing mix of algal and higher plant organic input.

3. The results of carbon isotope analysis of gas and crude oil samples

In the relationship diagram between carbon-13 isotope of Methane and Ethane (Fig.8a), the gas sample of the MT-1RX well is related to associated gas in the oil window phase. Meanwhile, gas of the HT-1X well was generated in the dry gas phase. The results of carbon isotope analysis of DST and RFT gas samples from wells such as 06-LT-OBS1, 06-LT-1XR, 06-LT-2X (1,662 - 1,672m), 06-LT-1XR and 06-LT-2X (1,682m) show that gas samples of 06-LT-1XR and GK 06-LT-1XR wells are a mix of petroleum gas and biogas. The presence of biogas in these gas samples suggests that the structures are open.

Gas of the 06-LT-OBS1, KCT-1X ST1 and LT- 1X wells are related to the oil window phase. On the other hand, gas of the HT-1X well was generated in the dry gas phase.

The carbon isotope analysis of gas samples shows various things as follows:

- There were two sources of gas:

+ Gas was generated in the transformation processes of organic matter in source rocks;

+ Biogas.

- The hydrocarbon accumulation potential of reservoirs.

In the exploration process, evaluating the generating potential is one of the three basic factors to study about petroleum systems. In the exploitation, identifying the interformation capacity in product fields plays an important role in modeling the fields. Moreover, the results of carbon-13 isotope analysis of oil samples are useful data for classifying the origin of oils.



Fig.5b. The results of GCMS analysis of crude oil sample (RFT, 3069.5m) in Middle Miocene reservoir at the 05-2-KCT-1X well [2]



Fig.6a. The results of GCMS analysis of condensate sample (MDT#2 - 2,956m) in Middle Miocene reservoir at the 05-2-HT-1X well [2]



Fig.6b. The results of GCMS analysis of condensate sample (MDT#2 - 3,195.8m) in Middle Miocene reservoir at the 05-2-HT-1X well [2]



Fig.6d. The results of GCMS analysis of condensate sample (MDT#2 - 3,617m) in Early Miocene reservoir at the 05-2-HT-1X well [2]



Fig.6e. The results of GCMS analysis of condensate sample (MDT#2 - 3,765m) in Early Miocene reservoir at the 05-2-HT-1X well [2]

Fig.8b (the relationship diagram between the Pr/Phy ratio from GC analysis of saturated hydrocarbon fraction and the results of carbon-13 isotope analysis of crude oils) shows the lacustrine origin of organic matter in source rocks generating oils at the NT-1RX well. Source rocks which generated oils at the HT-2X, MT-1X and KCT-1X wells contained mainly higher plant organic input. This is appropriate to the results of GCMS analysis listed above.

4. Conclusions

- The results of GC and GCMS analysis of crude oil and condensate samples as well as carbon-13 isotope of oil and gas samples are reliable data. The combination of these data is a useful tool to evaluate the origin of organic matter of oil and gas.

- The analysis results, namely GC component of groups, the characteristics of oils and condensates and bio-markers show that oils and condensates found in the Moc Tinh and Hai Thach structures have the following features:

+ Oils and condensates in the Moc Tinh and Hai Thach structures were generated from highly mature source rocks which contained mainly organic matter from higher plants and were deposited and degraded under oxidising and weakly reducing conditions.

+ However, all 5 oil samples in the Hai Thach structure have signals of marine organic input. This means that they originated in fluvio-deltaic source rocks.



Fig.7. The results of GCMS analysis of crude oil sample (RFT#2 - 2,864m) in Early Miocene reservoir at the 05-2-NT-1X well [2]



Fig.8b. Classifying the origin of oil by carbon isotope data in Blocks 05-2 and 05-3

- Oils in the Nguyet Thach structure were generated from lower maturity source rocks which contained a mix



Fig.8a. Classifying origin of gas by the relationship diagram between carbon isotope of methane and methane contents in Blocks 05-2, 05-3 and surrounding areas

of lacustrine and higher plant organic matter, deposited and degraded under reducing and weakly oxidising conditions. The RFT oil sample at 2,864m was destroyed.

- Crude oils and condensates in Blocks 05-2 and 05-3 were generated from Tertiary aged source rocks.

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SOME COMMENTS ON THE POSSIBILITY OF MIDDLE/LATE MIOCENE - PLIOCENE STRATIGRAPHIC TRAPS IN THE CENTRE OF NAM CON SON BASIN

Pham Thanh Liem¹, Phan Giang Long¹, Le Hai An² ¹Vietnam Oil and Gas Group ²Hanoi University of Mining and Geology

Summary

Stratigraphic traps are a new exploration target in Vietnam. To comment upon the possible existence of stratigraphic traps in the Nam Con Son basin and, in particular, of a Middle/Late Miocene - Pliocene stratigraphic trap in the centre of the Nam Con Son basin requires research on: (i) the regional tectonics which affected the existence and development of deposits; (ii) the sedimentary facies and sedimentary environments; and (iii) the process of formation of turbidite fans in the deep-sea environment in the region.

The deep-sea sediments of the Middle/Late Miocene - Pliocene age in the Nam Con Son basin constitute one of the hydrocarbon plays that are being studied by geo-scientists and international companies active in the oil and gas domain. Here, the present authors wish to introduce a case study which is summarised on the basis of geological, geophysical and drilling data from the centre of the Nam Con Son basin, the views of domestic and foreign geo-scientists of the characteristics of turbidites and the ability of deep-sea sediments to exist as turbidites of the Middle/Late Miocene - Pliocene age in the centre of the Nam Con Son basin.

1. Contents

A synthesis of researches conducted by geoscientists worldwide on the depositional conditions of stratigraphic traps shows that there are 3 main factors that affected the formation of stratigraphic traps: tectonics, sedimentary environment, the rise and fall of sea level. Each of the above elements will be analysed, then the possibility of the existence of deep-sea sediments as turbidite deposits in the Middle/Late Miocene - Pliocene in the centre of Nam Con Son basin will be discussed.

Based on the characteristics of sedimentary environments, including sediment supply and sediment type, in the Nam Con Son basin, there are 3 possible kinds of stratigraphic traps as below:

- Stratigraphic traps in the continental environment (Oligocene);

- Stratigraphic traps in the coastal environment (Early - Middle Miocene);

- Stratigraphic traps in deep-sea environment (Middle/Late Miocene - Pliocene).

This paper will focus on analysing the characteristics

of the deep-sea stratigraphic trap of Middle/Late Miocene - Pliocene age, including its tectonic features, stratigraphy and sediment characteristics.

1.1. Tectonic characteristics

The main tectonic elements that affected the formation of stratigraphic traps in the Nam Con Son basin have been mentioned by Hoang Ngoc Dang in "*Geology* and Potential Petroleum Resources" [1] and in many of his research studies on the processes of deposition and development of sedimentary basins in Vietnam [2]. They can be summarised as follows (Fig.1):

- The influence of the East Sea spreading in the Southwest, toward the Nam Con Son basin during the early Miocene epoch;

- The subduction and the cessation of the East Sea spreading in the Middle Miocene epoch (15ma);

- The upwelling of asthenosphere causing uplift and erosion of the region (sub-aerial erosion) to create the Middle Miocene Unconformity (MMU).

After the second rift in late Middle Miocene times, forming the regional Middle Miocene Unconformity,



Fig.1. The tectonic elements in South - East Asia that affected the Nam Con Son basin [3]



Fig.2. Tectonic elements in the Nam Con Son basin (pre-rift, syn-rift and post-rift) [1]



tectonic activity throughout the region was quite stable, with the sea-level rising and floods prevailing on the entire basin area under the influence of a thermal subsidence regime. Most fault activities ended in Late Miocene times and the basin's boundary is almost uniform across the region [1, 2].

2. Sediment characteristics

Under the influence of the rapidly rising sea level, the source material transported from the West was deposited in the centre of the Nam Con Son basin with a high rate of sedimentation due to the increasing sea-floor depth, with the seabed topography changing suddenly... The deepsea deposits had been formed and preserved since a tectonic influence no longer existed.

The process of erosion, transport and accumulation in deep water in the period after the Middle Miocene Unconformity had formed in the Nam Con Son basin was mainly controlled by gravity flows caused by subsidence, sedimentary sliding and turbulent flow related to the shelf - slope.

Many studies show that the turbidite formed at the edge of the basin related to high density currents. Characteristically in turbidity flows the transported material (under the turbulent currents impact) can move a long distance on a slight slope (low angle), usually onlapping the slope or underlying highs and are discontinuous downlapping onto the underlying and sediments, with diversified sizes. The formation and characteristics of deep-sea sediments reflect the complex interactions between internal and external factors, including the rise and fall of sealevel, tectonic processes at the edge of the basin, the type of the source material and the speed of supply.

2.1. Stratigraphy and sedimentary environments

In the sedimentary description, the turbidite of the Nam Con Son formation $(N_1^3 ncs)$ is distributed in the central part of the Nam Con Son basin, with the sand component being moderately to well-rounded and sorted, containing marine fossils and glauconite.

According to the characteristics of these sediments and the palaeo-biology, the Nam Con Son formation generally formed in a deep-sea environment, on the inner-shelf of the western part and the middle - outer-shelf of the eastern part of the basin [4].

The depositional turbidite in the deep-sea environment shows the net pay distribution sets with different characteristics for each area (inner-fan and outer-fan) (Fig.4) [9].

2.2. Flow

The unstable flow, with high density and low transport efficiency, tends to result in increased accumulation of sand close to the shelf edge. The turbidity current creates well sorted sand bodies, which are reworked materials from coastal plains or redeposited remains of the screened shelf-edge, and usually deposited at downslope. The lack of fine-grained material causes a reduction of fluid density, buoyancy of the liquid and the turbulent flow so that the sand cannot be transported far from sediment

source supply. The mechanism of turbidite formation and the sediment source supply is shown in Fig.5.

To consider the influence of ocean currents in the transportation and re-distribution of deep-sea sediments in the Middle/Late Miocene - Pliocene age in the Nam Con Son basin from all aspects requires very detailed analysis of the paleo-geographic environment in order to restore the slope-shelf's topography during this period, and analysis of the influencing factors related to ancient flows.

2.3. Sediment sources

A deep-sea turbidite can be divided into smaller components based on the volume, the particle size, and the characteristics of the sediment source supply. The sediment source study may indicate the volume and the general characteristics of the deep-sea fan, whereas the sediment supply will indicate the overall shape and distribution of the turbidite fan. The particle size reflects the composition of sediment source supply and the sedimentary processes: transportation efficiency, layered sedimentation, and the distribution of coarse and fine grain sizes in turbidite fans. In fact, the channels formed will develop into a lobe when the slope-angle becomes reduced and reduces the flow rate, and/or a change of flow direction (degree of bending of the flow). Sediments that filled up the channel/lobe overflow and form new channels in the slope/deeper shelf. While the sand is deposited in the lower region, the lighter clay/shale then settles on the top of it (Fig.6).

As mentioned above, the process of erosion, transport and sedimentation in the deep-water environment in the period after MMU in the Nam Con Son basin is mainly controlled by gravity flow, which causes the slumping or sliding processes of shale and silt. Deposition changed from a continental environment to a shallow marine environment (at the West - Southwest of the basin) and the transported materials, which are well sorted grain sands from coastal plains or redeposited remains of the screened shelf-edge, usually accumulate at the foot of the slope. However, to confirm the fan's size as well as the composition and the grain size of the sand body in the fan



Fig.4. Turbidite thickness distribution [9]





Fig.6. Turbidite deep-sea sediments system [10, 11]

based on the seismic analysis only (seismic interpretation attributes) needs further studies of the well-logs and the analysis of cores.

3. The rise and fall of sea level

There is evidence that changes in relative sea level can affect turbidite formation and the development of deep-sea fans in the stratigraphic column of the Nam Con Son basin. Many examples in the world (Indus fan, Bay of Bengal fan, Mississippi and Amazon fan) show that the amount of continental sedimentary clastics suddenly fell when the sea level rose during the Holocene. Most of the hydrocarbon-bearing fans are related to the fall of relative sea level as lower sea levels have a profound impact on erosion of the continental, sea-edge and deep-sea sediments. Lowstand submarine fans or basin floor fans are believed to be systems with a high ratio of sand/clay; this formed during the time the river system rejuvenated with large transportation rates due to slope increases (related to the period of lower sea level). During this time, sea-shelf sediments were mainly material which was transported through, with the coarse-sand supply settling on the outer-shelf. This gradual shift towards the sea is related to the fall of relative sea levels, detected by the onlap backwards to the

sea on the seismic records, the change of parasequence deposition type on the shelf or slope, and the sudden change from the deep-sea mudstone to clean sand on the well-logs' curve.

The joint study between Talisman and EPC/VPI (Fig.7) shows the sea-level fluctuations in the Oligocene -Miocene epochs and the effects of sea level on Oligocene -Miocene sediments in general and the deep-sea turbidite of the Middle/Late Miocene - Pliocene age (late stage of the syn-rift to post-rift) in the Nam Con Son basin in particular.

Based on the geochemical analysis, VPI has created a sedimentologic model of the Nam Con Son basin following the rise and fall of relative sea levels and the environmental characteristics of the sediments (Fig.8) [4].

Through the specific examples of the results of the seismic attribute analysis based on the 3D PSTM seismic cube of Block 04-1 (Fig.9), the characteristics of the stratigraphic trap of the Middle/Late Miocene - Pliocene age in the centre of the Nam Con Son basin should be considered as follows:

- Turbidite fan of the Middle/Late Miocene - Pliocene age deposited in the deep-sea environment, in the centre of the Nam Con Son basin is located at the shelf edge, especially in the transition zone between the inner-shelf



Fig.7. The rise and fall of relative sea level in the Nam Con Son basin [4]

and the outer-shelf where the topography of the sea-bed changes abruptly. Source material was transported from the mainland (western) and with the sea-bed's sudden change of depth the sediments were deposited from a turbulent flow as a turbidite fan...

- Tectonic activity in the region was quite stable during this period, the sea level increased and floods prevailed on the entire area of the Nam Con Son basin. Tectonic regime of thermal subsidence does not much affect the formation and preservation of the turbidite fans.

- During the Tertiary era the rise and fall of global sea level in Southeast Asia in general, and the Nam Con Son basin in particular, is very limited, so it is not the main factor to influence the stratigraphic trap at this time.

- The deep-sea sedimentary environment of turbidite is established, based on the quantitative well logs analysis. The basin floor fans (inner fan and outer fan) which are distributed in the central region of the Nam Con Son basin formed in the shelf-slope and shelf-edges.

- The turbidite (coarse sand) is located at the base of the shelf-slope transition zone, as the inner-fan with medium to well rounded and sorted sand, with the net pay of a few tens of cm to 1m (reflected in the well-log analysis).

- In order to delineate the turbidite within the study area, it is necessary to use the analysis of seismic attributes as a tool, combined with Spectral Decomposition and well-log analysis, using an Artificial Neural Network (ANN).



Fig.8. Sea level variation in the North and South parts of the Nam Con Son basin [4]



Fig.9. The images of turbidites based on the results of seismic attribute analysis using ANN in Block 04-1 (top of Late Miocene)

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APPLICATIONS OF THE SEISMIC ATTRIBUTES CUBE IN THE CUU LONG BASIN, OFFSHORE VIETNAM

Nguyen Thu Huyen, Nguyen Trung Hieu, Tran Ngoc Minh Vietnam Petroleum Institute

Abstract

Seismic attributes are a powerful aid to seismic interpretation. They allow us to interprete faults and channels, recognise the depositional environment and unravel the structural deformation history more rapidly. This paper discusses several examples of seismic attributes as CTC and ESP from the Cuu Long basin offshore Vietnam.

Seismic coherency (CTC) is a measure of the similarity between seismic traces. Coherency data play an important role in the delineation of structural and stratigraphic features by enhancing the images seen on the conventional 3D seismic data. A coherency cube was utilised to enable a quick interpretation of the structural framework of an area of a block in the Cuu Long basin. In particular CTC attribute slices near the surface of basement, together with fault interpretation of seismic data, could indicate the presence of possible smaller localised faults the basement surface of C and VT structures.

Event Similarity Prediction (ESP) is a seismic discontinuity volume attribute. Seismic discontinuity attributes quantify the degree to which neighboring seismic traces vary from each other. They detect abrupt lateral changes in seismic data character caused by faults, diapirs and stratigraphy. ESP data were used to assist prediction of the distribution of faults/fractures zone inside the CNV basement area. These examples illustrate how CTC and ESP data can be used in exploration to improve the imaging of geological features.

1. Introduction

A seismic attribute is any measure of seismic data that helps us visually enhance or quantify features of interpretation interest. A good seismic attribute is either directly sensitive to the desired geologic feature or reservoir property of interest, or allows us to define the structural or depositional environment and thereby enables us to infer some features or properties of interest. First introduced in the early 1970s, seismic attributes are now used widely for lithological and petrophysical prediction of reservoir properties [1 - 3].

The Cuu Long basin is one of the potential basins of Vietnam where petroleum activity is active. From the end of the1990s, seismic attribute analysis started to be applied to the exploration technology in this basin, primarily because the industry had embraced 3D technology - which was by far the most successful new exploration technology [4, 5]. The single most important contribution in making drilling decisions at that time was the concept of 3D attribute extractions. Finally it was possible to compute attributes for full 3D volumes and examine features of interest in their three dimensional perspective. By animating through those volumes, geosientists could quickly interpret variations in structural and stratigraphic style from seismic line to seismic line, and could quickly link subtle variations in seismic lines to their corresponding attributes.

With the Cuu Long basin where 3D seismic surveys had been done and were being conducted annually, good 3D interpretation workflows on interative workstations were being perfected. Complex trace analysis was performed on full 3D seismic volumes and used in the interpretations. However, most 3D seismic interpretation was performed on vertical inlines and crosslines and then projected onto time slice. Although that worked well, it led to ambiguities in the lateral resolution of faults, especially where faults joined together, crossed, or simply ended as a result of changes in geologic stress [1 - 3]. In order to resolve these events, the method of revealing fault surfaces within 3D volume for area where no fault reflections have been apply, these are CTC and ESP method. By using CTC and ESP, we could indicate the presence of possible smaller localised faults near basement surface of structures in the Cuu Long basin (structure C and VT) as well as predict the distribution of faults/fractures zones inside the CNV basement area.

2. Example of Applying Seismic Coherency into Block 17

The area of CTC application is located in the Western part of the Cuu Long basin (Fig.1). We used two 3D seismic surveys: 460km² of 3D survey acquired by PGS and processed by Golden Pacific Group and 152km² 3D survey carried out by Enterprise Oil Exploration Limited in 1996. In the area, the sea-floor varies from 40 - 55m water depth, except above some recent coral reefs (one of them rises up near sea level, which deranged the acquisition in the Northern part of the study area). The area is close to the Dragon oil field group, where oil is produced from Early Miocene Sandstone and Pre-Tertiary fractured basement. Our objective was to interpret two 3D seismic data cubes including attribute processing and analysis in combination with well data to describe the geometry of potential reservoirs and to evaluate their prospectivity.

CTC computes local trace to trace dissimilarity. These dissimilarity measurements yield the visual identification



Area applying CTCArea applying ESPFig.1. Location map of the applications seismic attributes areas

of such features as faults, facies changes, and other geologic patterns. Faults and stratigraphic changes will often standout as prominent anomalies in otherwise homogenous data. CTC processing converts normal seismic data into CTC attributes by measuring lateral data similarity through correlation and semblance techniques.

We could view and interpret CTC attributes just as normal seismic amplitudes. The advantage of the CTC data is that contrasts between similar and dissimilar data are much more apparent. Conceptually, we would expect high similarity values from trace to trace when the geology is flat and continuous, lower values when it is dipping and continuous, and anomalous values when it is discontinuous. This is very useful if we try to pick faults or delineate stratigraphic changes [1 - 3]. We are using available Landmark software Postack-ESP3D to produce CTC attributes, and use SeisWorks 3D for analysis of CTC attributes with the purpose to predict the development of the fracture system of near surface of basement in the area around the "Cam" and "Vai Thieu" structures.

Not all the 3D seismic data cube is selected but, a subset volume has been chosen around the "Cam" and "Vai Thieu" structures in order to reduce the processing time. Fig.2 shows the interpreted seismic section through these structures.

CTC attribute cubes of selected areas were generated and used for analysis. Fig.3 and 4 show CTC attribute slices at 2400ms and 1860ms near surface of basement for Cam and Vai Thieu structures respectively. In these slices, the dark bands show areas of low similarity or dissimilarity and we would expect to see in these areas the possible presence of faulting. The white zones show the trace to trace similarity. Analyzing several CTC attribute slices near the surface of basement, together with fault interpretation of seismic data, we would expect the presence of possible smaller localised faults near the basement surface of Cam and Vai Thieu structures. This indicates the possible presence of fractures in the near surface of basement in the Cam and Vai Thieu structures (Fig.5).

3. Example of Applying ESP into Block 09

This example was done by using ESP (Even Similarity Prediction) processing and analysis of approximately 260km² data selected from 3D PSDM 2004 full stack volume acquired for Hoan Vu Joint Operating Company to study the distribution and intensity of faults and fractures within the basement rocks of Ca Ngu Vang (CNV)



Fig.2. Interpreted seismic section through VT (a) and Cam (b) structure



(a) (b) **Fig.3.** Cam area. Time slice (a) and CTC slice (b) at 2400ms



Fig.4. VT area. Time slice (a) and CTC slice (b) at 1860ms



Fig.5. Coherency map along basement horizon overlaid fault systems

field in the block 9-2 (Fig.1) as well as the possible reservoir quality distribution regarding the faults and fractures distribution.

We used available Landmark software "Post-stack-Structure Cube" to produce and analyse ESP attributes with the purpose of studying the distribution of fault/fractures systems [1 - 3].

Structure cube is a seismic discontinuity volume attribute. Its primary application is to serve the purpose: facilitate 3D seismic interpretation by highlighting faults, channels, and other geologic features, especially in a time-slice view. Seismic discontinuity attributes quantify the degree to which neighboring seismic traces vary from each other. They detect abrupt lateral changes in seismic data character caused by faults, diapirs and stratigraphy. Structure Cube compares all the traces in an analysis window with the average trace of the window to compute seismic discontinuity. The comparison is accomplished through weighted correlations, which are sensitive to the relative magnitude of the trace amplitude values, unlike standard correlation. It scales the output data to be in the range of 0 - 100, with 0 being perfectly continuous, and 100 being perfectly discontinuous. For interpretation, one can view and interpret ESP attributes just as one would normal seismic amplitudes. Figures 6 and 7 show the ESP results processed from PSDM 2004 full stack data. In order to interpret the results, in these pictures, the light bands indicating the lowest values (near 0) refer to "being perfectly continuous" and the dark-bands indicating higher values (near 100) refer to "being perfectly discontinuous". To recognise features such as faults we can use the darkbands for interpretation.







Fig.7. ESP depth slice at 4152m (a) and at 4256m (b) with faulst/fractures interpretation

4. Conclusions

The association of attributes with 3D seismic data introduced new life into attribute analysis, moving it away from seismic stratigraphy and toward exploitation and reservoir characterisation. Seismic attribute as CTC and ESP has a important role in improving the confidence of interpretation by improving the imaging of structural fabric and stratigraphic features. They are of relatively new application and are to be integrated with other technologies to provide additional insights. Two examples have been discussed showing the integration of seismic attribute cubes in an exploration setting to improve the imaging of geological features ranging in scale from reservoir barriers to faults/fractures. Applications of the attribute cube in the Cuu Long basin enables geocientists to continue to extract more and more information from seismic data.

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DEVELOPMENT AND APPLICATION OF MULTI-WAVE UVP AND PIV MEASUREMENT METHODS TO MEASUREMENTS OF SINGLE-PHASE AND BUBBLY TWO-PHASE FLOWS IN A VERTICAL PIPE

Nguyen Tat Thang¹, Duong Ngoc Hai^{1, 2}, Hiroshige Kikura³

¹Institute of Mechanics, Vietnam Academy of Science and Technology, Vietnam ²University of Engineering and Technology, National University of Hanoi, Vietnam ³Tokyo Institute of Technology, Japan

Summary

Two advanced measurement methods, i.e. multi-wave UVP (ultrasonic velocity profile) and PIV (particle image velocimetry) methods have been developed and applied to the measurement of single-phase and bubbly two-phase flows in a vertical pipe. These are non-intrusive measurement methods which are able to measure the instantaneous velocity profile and 2D velocity field. Thanks to the instantaneously measured velocity profile of single-phase and two-phase flows the characteristics and behaviour of single-phase and two-phase flows can be clarified. Relevant flow parameters can be obtained. The multi-wave UVP method, which is highly compact and able to work with opaque fluids and containers, is expected to be applied to the practical measurement of instantaneous velocity profiles, flow rate and local void fraction, etc., in industrial piping systems in operations such as oil and gas two-phase flows with the presence of deposition, crystalisation and corrosion. Therefore, it could be a very useful measurement tool in the oil and gas and power generating industries.

1. Introduction

Single-phase and two-phase flows play important roles in many industrial, technical and engineering systems. Some of the many examples are oil and gas flows in oil and gas and petroleum industry, water-steam flows in piping systems in heat removing systems, and in steam generators in energy systems. In such systems, the characteristics and structures of flows have direct correlations with many scientific and technical problems such as proper, optimised and safe operation and control of the whole system. Hence thorough understanding of single-phase and two-phase flows has very important practical implications. In addition to the single case of fully developed laminar flow in pipes, which has a beautiful analytical expression of parabolic radial velocity distribution, all of other cases are turbulent single-phase and two-phase flows whose behavior and structures vary a lot depending on the flow regime and flow geometry. There is no universal analytical expression for these flows. Therefore study of the characteristics and structures of single-phase and two-phase flows in pipes still attracts much attention [1, 2].

One of the most important parameters required for

study of single-phase and two-phase flows in piping systems is instantaneous velocity distribution of liquid (in single-phase flow) and of liquid and gas (in two-phase flow). Methods for calculation of pipe flow velocity include numerical simulation and experimental measurement. Numerical simulation based on solution of mathematical models using PC has gradually improved due to enhanced models and computing power. It has advantages such as being possible for a wide range of flow parameters. However numerical models usually have to adopt various kinds of assumptions including approximations, averaging and model closures. Especially, two-phase flow models currently have to rely on experimental measurement data for many kinds of correlations between the liquid and gas phases. As a result, experimental measurement of single-phase and two-phase flows plays a tremendously important part. A major advantage of the experimental study of pipe flow is the possibility to confirm measured data directly. Exact flow parameters can be obtained using measurement techniques. The data can be used not only for study of flow, for calibration and validation of numerical models but also as data for boundary conditions of the models. Therefore properly measured data is highly important to build up knowledge and data banks

for single-phase and two-phase flows, especially flows in industrial piping systems [1, 2].

Experimental methods for measurement of velocity of liquid flow can be classified into intrusive methods and non-intrusive methods. Intrusive methods all involve insertion of measuring probes into the flow field for measurement. Some typical examples are hot-wire and hot-film anemometries, wire mesh tomography and pitot tubes the intrusive effects of such methods disturb the flow field and cannot be neglected. In addition, experimental setup tends to be more complicated. Interuption of flow in operation may be required for the deployment of measuring probes. Therefore measurement of systems in operation is difficult. On the other hand, non-intrusive methods are always more attractive taking into account the avoidance of interference between the measuring probes and liquid flow. Popular examples of non-instrusive measurement methods are laser Doppler anemometry (LDA), ultrasonic velocity profiler, particle image velocimetry and nuclear magnetic resonance (NMR). The LDA method is a point-wise measurement method which needs an optical window for the operation of a laser beam. The UVP method is a velocity profile measurement method using ultrasound and signal processing. It is a non-intrusive and non-contact measurement method since ultrasound can penetrate many materials. Similar applications of ultrasound are very well known in the medical field. The PIV method is based on the statistical processing of particle images of the flow field. It can produce 2D and 3D velocity fields of flows with very high temporal and spatial resolutions. However it requires an optical windows for the exposure of the flow field into a laser sheet. NMR is also another non-intrusive and non-contact measurement method. It can obtain results in 2D and/or 3D. However the NMR method uses bulky hardware for safety protection and usually needs many operators for measurement. Therefore it is not suitable for measurement at small experimental sites, and in industrial on-site conditions. Taking into account the characteristics of the measurement methods listed above, the UVP and PIV methods would be superior to other methods for measurement of singlephase and two-phase flows in a vertical pipe.

The principle of the UVP method, which was originally developed for single-phase flow measurement, is extracting velocity information carried in ultrasound when it is reflected from moving objects. Therefore this method uses an ultrasonic transducer (TDX) to emit ultrasonic pulses into the flow field. In order to do that, a

pulser/receiver (P/R) is used to generate electrical pulses to excite the piezoelectric element of the TDX. Ultrasound is reflected from particles seeded in the flow. Echo sound is then received and converted into a voltage signal by the same piezoelectric element of the TDX. The electrical signal is filtered and amplified by the P/R. An analog to digital converter (ADC) is used to digitise the voltage signal and transfer it to a PC. Signal processing techniques are applied to the received signal to extract flow velocity along the sound path. Since ultrasound can penetrate various materials and liquids, the UVP method can be used with opaque fluids and non-transparent containers. On the other hand, the PIV method is basically based on optical principles to obtain particle images of the flow field, and statistical methods for image processing. To capture particle images, the flow field is usually exposed to a laser (or light) sheet. Image processing techniques are applied to the recorded images to extract the velocity of the flow. Therefore the PIV method can provide detailed 2D or 3D velocity fields. However it must use an optical window for imaging the flow field. As a result, it can not be applied to opaque liquids and containers which are very often encountered in flows in industrial and engineering piping systems [3].

The UVP and PIV methods were originally developed for single-phase flow measurement. Application of these methods to two-phase flow measurement is not straightforward. In gas-liquid two-phase flows, which are very often encountered in reality, both ultrasound and light are scattered strongly at gas-liquid interfaces due to very high difference of acoustic impedance between liquid and gas. Received signals or data include information of both liguid and gas. That is also a problem with the LDA method when it is applied to two-phase flow measurement. In order to obtain instantaneous velocity of each phase, phase separation methods are required to select liquid data and gas data separately, either at an early stage to selectively collect data of each phase only or at a later stage to properly select the velocity of liquid phase only and gas phase only. A combination of both ways is also possible. A common solution to this problem is using multi-wave (ultrasound in UVP method or light in PIV method). The principle is using an appropriate wave for measurement of each phase. When two waves are used, measurement of two-phase flow is possible [4].

In this study, we have developed two methods for measurement of single-phase and two-phase flow velocity in a vertical pipe. The first method uses two ultrasounds with different centre frequencies and is called the multiwave UVP method. This method can measure the instantaneous velocity profiles of each phase separately. For measurement of single-phase flow of liquid, this method works as the original UVP method in which only one frequency is used. The other method is a PIV method for only the gas phase in a two-phase flow. For single-phase flow measurement, the method works as normal PIV measurement. As part of the development of the measurement methods, test flow loops have been developed to confirm the validity of the two methods. For single-phase flow, instantaneous velocity profiles have been properly obtained for various Reynolds number (Re). For single-phase and bubbly two-phase flows, the multi-wave UVP method is confirmed using PIV data. Instantaneous velocity profiles of liquid and gas have been obtained for various liguid and gas superficial velocities. It is shown that multiwave the UVP method exhibits robustness in two-phase flow measurement both in research and in engineering. The measured result is used for further analysis of relevant parameters (e.g. flow rate, local void fraction etc.) of single-phase and two-phase flows. In addition, measurements can be extended to other cases of single-phase and bubbly two-phase flow in a pipe.

2. Measurement methods for single-phase flow

2.1. UVP method

There are two signal processing methods for the calculation of velocity including the ultrasonic Doppler method (UDM) which exploits the Doppler effect, and the ultrasonic time-domain cross-correlation method (UTDC) which is a statistical method for analysis of time series data, i.e. echo waves. The first method was developed early, and is suitable for early electronics [5]. It has been widely applied in commercial UVP systems. The principle of the Doppler method will be introduced and exploited in this study. The other method, which is UTDC, has recently been developed. Due to the complexity of correlation algorithm, it tends to be computationally expensive. Early applications of UTDC were usually for offline measurement. Readers who are interested in the UTDC method can refer to [6]. At present, there is another method for measurement of fluid flow of very low velocity. The method is known as the phase difference method and is under development [7]. As a result, details of the principle of the ultrasonic Doppler method will be presented below.

In the Doppler method, the received digitised signal is analysed to obtain the Doppler shift frequency $f_{d'}$ i.e.

the change in frequency between emitted ultrasound and the reflected one (Doppler effect). The Doppler shift frequency has a direct relationship with flow velocity as shown below:

$$f_{d} = 2f_0 \frac{v}{c} \tag{1}$$

Where:

v is the fluid velocity which has the direction in the sound path;

 $\boldsymbol{f}_{\scriptscriptstyle 0}$ is the central frequency of the transmitted ultrasound;

c is the sound speed in the working liquid.

The principle of the Doppler method is explained in Fig.1.

As shown in Fig.1, the TDX emits an ultrasonic pulse into the flow field and switches to the receiving mode to receive echo signals. The reflected ultrasound from all depths in front of the TDX surface is received. The sound speed in the working liquid is known a priori. The measurement channels (the positions along the sound path where velocity is measured) are specified according to the time when the echo signal is sampled for the calculation of velocity. This is also known as the range-gated time system. For example, in Fig.1, the echo signal from the enlarged position is the signal that is sampled from the time series of echo signal after a time delay Δt (with the time origin being the time when the TDX emits an ultrasound pulse).





The time delay is calculated according to Eq.(2) where Δx is assigned as desired in measurement setting:

$$\Delta t = 2\Delta x/c \tag{2}$$

Similarly for other measurement channels, signal processing of echoes from the channels provides Doppler shift frequencies at the channels and velocity of the channels are calculated straight forwards using Eq.(1). Since Doppler shift frequency is determined for the movement in the sound path direction, it can not be calculated for the movement in the direction perpendicular to that direction. As a result, in practical measurement, the TDX must be set inclined at an angle θ to the flow direction (Fig.1). Hence the flow velocity at a measurement channel is calculated using Eq.(3):

$$v = \frac{f_d}{f_0} \cdot \frac{c}{2\cos(\theta)} \tag{3}$$

Though each ultrasonic echo wave from a measurement channel does contain some information on the Doppler effect as a result of the movement of the fluid flow, it is currently impossible to extract Doppler shift frequency by using just only one echo wave which is corresponding to one emitted pulse. The reason is mainly due to the limitation of the speed of the digitiser. In a commercial UVP system, the numbers of echo waves from a measurement channel (corresponding to the number of emitted pulses) typically are 16, 32, 128, 256 and so on. These numbers are a power of 2 due to the fact that Fourier analysis is usually adopted in the signal processing to obtain Doppler shift frequency. Typical methods for the derivation of Doppler shift frequency are zero crossing method, Fourier analysis using FFT algorithm, auto-correlation etc. [4, 5, 8]. Due to its robustness, in this study, the auto-correlation technique is exploited.

In a typical UVP system, the ultrasonic pulses are emitted at a pulse repetition frequency F_{prf} and time duration $T_{prf} = 1/F_{prf}$, F_{prf} itself also implies the sampling frequency of the Doppler signal whose centre frequency is the Doppler shift f_d . These parameters set direct constrains on the maximum measurable velocity (v_{max}) and maximum measurable depth (P_{max}) of the UVP method due to the limitation expressed in the Nyquist sampling law (i.e. $f_d < F_{prf}/2$), and the limitation of the time duration set aside (between two emissions) for the TDX to hear echo signal (i.e. $T_{for receiving echo signal} < T_{prf}$). Such constraints are written in Eq.(4) and Eq.(5).

$$v_{\max} = \frac{F_{prf}}{2f_0} \cdot \frac{c}{2\cos(\theta)}$$
(4)

$$P_{\max} < \frac{c\cos\theta}{2F_{prf}} \tag{5}$$

2.2. PIV method

1

A schematic diagram of the PIV method is briefly explained in Fig.2. Similar to the UVP method, the PIV method needs seeding particles as reflectors of light. It is usually assumed that seeding particles of approximately equal density with water are used. Therefore their effect on flow dynamics is negligible. The flow field is then exposed at least twice, in an appropriately short time interval, to a light sheet (which is usually a laser sheet). The image of the exposed area of the flow field is captured using a camera (recently using digital cameras). The captured images are image pairs or single images. The displacement of the particles between two adjacent exposures is estimated using statistical methods and image pairs. Based on the particle displacement, the velocity field of the flow is determined [9].

For the estimation of velocity using particle images, the images are divided into rectangular areas called interrogation windows (Fig.3). Statistical methods such as auto-correlation or cross-correlation are applied to the interrogation windows to determine particle displacement between two exposures.



Fig.2. Schematic diagram of the PIV method [9]



Fig.3. Interrogation windows and single exposure images [8]

Based on the displacement calculated for each interrogation window, spatially averaged (in the interrogation window) flow velocity of the windows is calculated [9]. Particle images are usually stored in gray scale images for the analysis of particle displacement. Currently, in a typical PIV system, the statistical method used is the estimation of the correlation coefficient of the time series data which are composed of two dimensional gray scale images. It is possible to calculate the correlation coefficient directly using particle images. However, very often, the correlation coefficient is calculated indirectly by adopting Fourier transform and Wiener-Khinchin theorem (Fig.4). The Wiener-Khinchin theorem states that Fourier transform of the auto-correlation function R_{μ} and the power spectral function $|\hat{I}(r_y, r_y)|^2$ of the light intensity field $I(x, x_y)$ y) are Fourier transform of each other [9]. In more detail, in the case of single exposure/double frame images as shown in Fig.4, the correlation function is written as:

$$R_{II}(x, y) = \sum_{i=-K}^{K} \sum_{j=-L}^{L} I(i, j) I'(i + x, j + y)$$
(6)

Statistically, the correlation function provides essential information on the similarity of the distribution of the light intensity (i.e. the distribution of particles) in I and I' corresponding to each movement in *x* and *y*. Therefore the maximum of the correlation implies the real displacement of the particles between two adjacent images. As a result, flow velocity is inferred. Direct calculation of the correlation function is extremely time consuming. As a result, indirect calculation based on Fourier transform is used in most of the current commercial and scientific PIV systems. The algorithm of the displacement estimation, which is based on Fourier transform, is shown in Fig.5.

In addition to the application of Fourier transform, interpolation methods are exploited for the optimal esti-



Fig.4. Schematic diagram of the displacement estimation method using Fourier transform [9]



Fig.5. Schematic diagram of the algorithm of the displacement estimation based on Fourier transform (figure from [9])

mation of the maximum of the correlation function of the particle displacement.

These methods are usually called sub-pixel estimation of particle displacement [9].

3. Development of measurement methods for twophase flow

3.1. Multi-wave UVP method

When conventional TDX and UVP methods are applied to measurement of single-phase flow, ultrasound is reflected by only seeding particles in the flow field. Only one central ultrasonic frequency f_o is used for measurement of liquid velocity. If the method is applied to two-phase flow measurements, ultrasound gets reflected from different reflectors including: seeding micro particles and interfaces between phases (e.g. liquid and gas interfaces). If only one TDX with one central frequency is used, it is difficult to classify the echo sounds reflected from different objects. Therefore it is very hard to tell which one is liquid data and which one is gas data. Moreover, if the interfaces between phases are large (compared with the cross sectional area of the ultrasonic beam), ultrasound can not go beyond the interface, i.e. it is blocked at the interface.

In the UVP method, measurement volume is a cylindrical shape specified by the ultrasonic beam diameter and the number of wave cycles in one emitted pulse. The thickness of the measurement volume is defined as $N\lambda/2$ where N is the number of wave cycle of one emitted pulse and λ is the wavelength. The measurement volume size has a relationship to the measured result. When different sizes are used, particular phase velocity in multi-phase flows can be measured. Experiments [11] showed that, if an ultrasonic beam of 2MHz centre fre-

> quency, 10mm diameter is used to measure bubbly flow whose bubble size is of an order of several millimeters, measured results mostly show bubble velocity. On the other hand, with the same flow conditions, if an ultrasonic beam of 8MHz centre frequency, 3mm beam diameter, is used, measured results exhibit mostly liquid velocity. As a result, the idea of using multi-wave ultrasounds for the measurement of two-phase flow has been proposed. If the TDX is possible to emit and receive two

ultrasounds of 2MHz and 8MHz frequencies simultaneously at the same position, phase velocities of bubbly flows can be measured. Based on this idea, the principle of multi-wave TDX and multi-wave UVP methods for the measurement of bubbly two-phase flows is devised and is shown below.

3.1.1. Multi-wave TDX

A multi-wave ultrasonic TDX is composed of two piezoelectric elements including 2MHz and 8MHz frequencies. Fig.6 shows a schematic configuration of a multi-wave TDX which can simultaneously emit and receive ultrasound of 2MHz and 8MHz frequencies independently at the same position for the measurement of bubbly two-phase flows.

The 8MHz frequency element is a cylindrical shape with diameter 3mm located at the centre and surrounded by the 2MHz frequency element which has an annular shape. Each element is controlled independently and simultaneously by two synchronised P/Rs. This uniquely designed multi-wave TDX enables the emission and reception of ultrasounds of 2MHz and 8MHz frequencies simultaneously and independently at the same position for bubbly flow measurement.

3.1.2. Pulser/receiver and data acquisition unit

Usually, the ultrasonic pulsed Doppler method is used with a tone burst P/R for the excitation of ultrasonic TDX and for reception of echo sound. However, we have proved that, by using low-cost, popular spike P/Rs in ultrasonic testing industry, measurement of instantaneous velocity profiles using a pulsed Doppler method is also possible [12]. Therefore, two spike P/Rs, which were synchronised, have been used to excite two piezoelectric elements of the multi-wave TDX simultaneously. After emission, reception of echo sound of both frequencies occurs at the same time. The captured signals of 2 and



Fig.6. A schematic configuration of multi-wave ultrasonic TDX

8MHz frequencies are converted into voltage signals by the corresponding piezoelectric elements respectively. Voltage signals are then amplified and filtered by the P/ Rs. For data digitisation and acquisition, a two channel, high speed ADC is used to connect with the two P/Rs. It is also synchronised with the P/Rs for data acquisition. Digitised signals of both channels are transferred into a PC for online signal processing to obtain instantaneous velocity profiles of two-phase flow.

3.1.3. Discrimination of gas phase from liquid phase

In two-phase flow, ultrasound is back scattered by both seeding particles and gas-liquid interfaces. Since the echo signal from gas-liquid interfaces is much stronger than that from seeding particles, with an appropriate small gain setting for the 2MHz TDX the 2MHz echo signal will contain only signal reflected from bubble surfaces. Therefore, it is assured that the 2MHz signal includes only bubble data. Though, it has been confirmed that measured data of 8MHz channel mostly includes liquid velocity, 8MHz frequency signal always includes bubble data if bubbles cross the 8MHz sound beam. Therefore, bubble data must be eliminated from liquid data to obtain velocity profiles of only liquid. For bubble data elimination, the relation between relative positions of bubbles and ultrasonic beams are all considered including the following cases (Fig.7):

- No bubble detected by both frequencies,
- One bubble crossing 2MHz beam,
- One bubble crossing 8MHz beam.

In pattern a), only liquid velocity profile is measured by 8MHz frequency. There is no bubble velocity (no bubble data) along the measurement line of 2MHz frequency. If one bubble crosses the 2MHz ultrasonic beam, case b), the bubble velocity is measured by 2MHz frequency; the liquid velocity is measured by 8MHz frequency at the same time (without bubble data since a bubble does not cross the 8MHz beam). When the bubble crosses the 8MHz beam, case c), both frequencies will measure bubble velocity. In this case, 8MHz frequency measures both liquid and bubble velocities. However, 8MHz ultrasound is obstructed by the bubble and the echo signal coming from the area behind the bubble must be considered as noise. As a result, the measured velocity behind the bubble needs to be deleted (Fig.7). With the proposed algorithm for phase velocity selection, instantaneous velocity profiles of both liquid and bubble can be measured



Fig.7. Relative positions of a bubble and ultrasound beams (up) and corresponding selection algorithms of valid liquid velocity (down)

simultaneously along the measurement line. Using this method, relative velocity between bubble and liquid is no longer needed for the phase separation.

3.2. PIV method for bubbly flow

Similar to the application of the UVP method to measurements of bubbly two-phase flows, when PIV is applied to measurement of two-phase flows, light is reflected from both seeding micro particles and phase interfaces. Reflected light from phase interfaces is usually much stronger than that from seeding particles. Therefore the influence of reflected light from phase interfaces on that from seeding particles surrounding the interface is very considerable. This effect makes it extremely difficult to distinguish between areas of fluid and gas surrounding the interface. Generally, the solution to this problem is using fluorescence as seeding particles for fluid phase. Moreover, there are two cameras, each using a specific optical filter corresponding to lights of original wave length and of fluorescence emission. This means that one camera will capture only reflected light with original wave length from phase interfaces. The other camera will capture only light emitted from fluorescence with wavelength changed when fluorescence is exposed to a laser light sheet. As a result, signals from liquid and gas phases are separated completely. Then PIV analysis can be carried out for each phase separately [13].

Due to the limitation of the hardware including optical filters and fluorescence, in our method, we measure gas phase only. Images of bubbly two-phase flow without seeding particles are captured in the same way as in the conventional PIV method. The thickness of the laser light sheet is adjusted to be about the same as the average bubble diameter. This reduces excessive light reflected from gas-liquid interfaces on surrounding areas. By using an appropriate digital image processing algorithm and an appropriate size of interrogation windows, instantaneous velocity field of gas phase can be derived. Using this method, it is possible to confirm bubble velocity calculated by the PIV method directly using bubble images. Statistically, it is possible to confirm measured data of the multi-wave UVP method by using the PIV method for two-phase flow.

4. Single-phase and two-phase flow experimental apparatus

4.1. Experimental apparatus for measurement of singlephase and two-phase flows in vertical pipes

A schematic diagram of the experimental apparatus for measurement of single-phase and/or bubbly two-phase counter-current flows in vertical pipes is shown in Fig.8a. The apparatus is located in the Laboratory for Industrial and Environmental Fluid Dynamics, Institute of Mechanics, Vietnam Academy of Science and Technology (VAST) and is shown in Fig.8b. Where: 1: Floor tank; 2: Water flow rate controlling valve; 3: Bubble generator nozzle; 4: Measurement pipe of I.D. 50mm for single-phase and two-phase flow study; 5: Overflow weir of floor tank; 6: Water circulation pump; 7: Bypass valves; 8: Pipe for water supply to the upper tank; 9: Overflow weir of the upper tank; 10: Upper overflow tank; 11: Water box for housing ultrasonic TDX; 12: Multi-wave TDX (Japan Probe Co. Ltd.); 13: Drainage pipe for overflow water down to floor tank; 14: Water drainage valves; 15: Main water drainage pipe; 16: Air compressor; 17: Main air flow rate controlling valve; 18: Air flow rate regulator system (Cole Parmer Co. Ltd.); 19: Float valve for air flow rate measurement (Tokyo Keiso Co. Ltd.); 20: Pulser/ Receiver of the UVP system (JSR Ultrasonics Co. Ltd.); 21: PC which houses ADC (National Instruments) and UVP software etc.; 22: Water flowmeter (Aichi Tokei Denki Co. Ltd.); 23: High power pulsed laser (Dantec Ltd.); 24: Camera (ordinary and high speed); 25: PC for controlling laser, camera, data logger and PIV software (Dantec DynamicStudio), etc; 26: Pipe for single-phase flow study (I.D. 26mm); 27: flow rate control valve of pipe 26.



Fig.8. Schematic diagram of the experimental apparatus



Fig.9. Typical two-phase flow regimes observed in the flow loop with 50mm I.D. pipe using a high speed camera (JVC Hybrid Camera GC-PX1, JVC Co. Ltd.) with frame rate 300Hz (a: bubbly flow; b: slug flow; c: annular flow)

As shown in Fig.8a, there are two test pipes: test pipe 4 with I.D. 50mm, 3m long and test pipe 26 with I.D. 26mm, 2.4m long. Both are made of transparent acrylic that facilitates UVP measurement and optical flow visualisation for the PIV method. Because the ratio of pipe length to pipe I.D. of large test pipe 4 is not enough for fully developed laminar flow, it is used for measurement of fully developed turbulent single-phase flow and two-phase flow. The small test pipe 26 has a higher ratio of pipe length to pipe I.D., it is used for measurement of fully developed laminar and turbulent single-phase flow. Water flow rates in the measurement pipes are controlled using needle-type valves at the outlet of the pipes (Fig.8a). The water tanks have overflow weirs to ensure a constant pressure difference between the two tanks. This experimental apparatus is used for measurement and study of single-phase and bubbly two-phase flows. In the case of bubbly counter-current flow, bubbles generated at the bottom part of the measurement pipe flow upwards. Water flows down from the upper tank to the floor tank solely under the effect of gravity. As a result, a bubbly counter-current flow is formed in the measurement

> pipe. Moreover, other flow regimes can also be created in the measurement pipe such as slug flow, annular flow, and churn flow etc. as shown in Fig.9. These gas-liquid two-phase flow regimes depend on the correlation between water flow rate and that of gas supplied into the measurement pipe. The main parameters of the experimental apparatus are shown in Table 1.

> Water flow rate in the measurement pipes is measured using a high precision turbine flowmeter located at the outlet of the pipe. The multiwave TDX is attached to the outside of the measurement pipes, and inclined at 45 degree angle to the main flow direction (the vertical direction). For making the same acoustic impedance with that of working fluid in the measurement pipe, a water box is used for housing the TDX. The TDX surface is always submerged in water. Moreover the water box is also used to eliminate the distortion effect caused by the curved wall of the measurement pipe. This is important for optical visualisation, flow observation and for PIV method. For small test pipe 26, TDX can be submerged in the lower tank for measurement. In other parts of the pipe, ultrasonic gel is used instead.

> In addition, it is possible to expand this experimental apparatus to cover also upward sin

Table 1. Main parameters of the experimental apparatus of single-phase and bubbly counter-current two-phase flows in a vertical pipe

| Type of flow | Single phase downward (laminar/turbulent) Bubbly counter-current flow (turbulent) |
|--|--|
| Material of test pipes | Transparent acrylic |
| Inner/outer diameter of pipe 4 | 50mm/60mm |
| Inner/outer diameter of pipe 26 | 26mm/30mm |
| Lengths of pipe no. 4/pipe no. 26 | 3m/2.4m |
| Range of liquid flow rate/Re of pipe 4 | 0 - 39.5L/min/0 - 20,000 |
| Air flow rate range (following the available range of the air flowmeter and the capacity of the air supply system) | 0 - 6L/min |
| Measurement, observation location | Along the measurement pipes |
| Two-phase flow regimes | bubbly/slug/annular |

gle-phase and/or co-current two-phase flows for a wider study of pipe flow.

4.2. Configuration of the UVP and PIV systems

4.2.1. The multi-wave UVP system

Fig.10 shows a schematic diagram of the advanced multi-wave UVP system in the Laboratory for Industrial and Environmental Fluid Dynamics, Institute of Mechanics, VAST.

Main parameters of the multi-wave UVP system are:

- Central frequencies: 8MHz (for liquid); 2MHz (for gas);
- F_{prf} of the P/Rs: 100Hz 5kHz;
- ADC: 2 channels, maximum sampling rate 100MHz.

UVP software.

4.2.2. The Dantec 3D-PIV system

An advanced Dantec 3D-PIV (stereo PIV) system has been adapted in the same laboratory for multi-purpose, fluid dynamics study. The system has the main features as shown below:

- High power, pulsed laser (Dantec Dynamics: Nd-YAG laser), synchronised with PC and camera;

- Maximum pulse power: 1,200mJ;
- Pulse emission time: 4ns;
- Laser wavelengths: 1,064 and 532nm;

- Cameras: high resolution 2,048 x 2,048, 8 bit grey scale;

- Maximum frame rate 15Hz;

Full, digital image processing software and 2D/3D-PIV softwares Dantec DynamicStudio.

4.3. Experimental conditions

Measurements were carried out in atmospheric pres-



Fig.10. Schematic configuration of the multi-wave UVP system for measurement of single-phase and two-phase flows



Fig.11. Schematic diagram of the Dantec 3D-PIV

sure, at room temperature. The multi-wave TDX was fixed at an inclined angle of 45 degrees to the main flow direction using a TDX holder. The thickness of pipe wall at the measurement locations was 1mm. Nylon powder (WS-200P, Daicel-Evonic, Ltd.) with average diameter of 80µm and density of 1.02g/cm³ was used as ultrasonic scatterers for measurement of liquid velocity and for PIV particle image. Measurement conditions are shown in Table 2 and 3.

5. Measurement results

5.1. Measurements of single-phase flows

Fig.12 shows the arrangement of the multi-wave TDX and the pipe flow under measurement.

The TDX is inclined at a 45 degree angle to the flow direction. Only 8MHz ultrasonic frequency is used for the measurement of liquid velocity in the pipe. The spatial

resolution in the sound path direction is 0.74mm which corresponds to a resolution of 0.523mm in the traverse direction which is perpendicular to the flow direction. For singlephase flow, only 8 MHz element of the multi-wave TDX is used. Measured results using only UVP method for fully developed laminar flow in the small diameter test pipe 26 are shown in Fig.13a.

Reynolds number in this case is around 1,400. As shown in Fig.13a, the measured result

agrees well with the analytical solution of parabolic profile of laminar pipe flow. However, close to the wall, velocities at some measurement channels are lower estimated by the UVP method as compared with the parabolic velocity profile of the exact solution. The same situation happened with measurement of turbulent pipe flow using the UVP method (Fig.13b). This is due to the effect of the measurement volume size of the UVP method [14]. Close to the wall, some measurement volumes comes in contact with the pipe wall. They include fixed part of pipe wall and outside liquid. This decreases the measured velocity in such volumes.

For measurement of fully developed turbulent pipe flow, either pipe, pipe 4 or pipe 26 could be used. In this study, the larger diameter pipe 4 was used. Measurements have been carried out using both UVP and PIV methods. Measured results are shown in Fig.13b. It can be seen that the UVP result in this case also shows the wall effect while the PIV result is better close to the wall. In addition, a typical particle image and a velocity field calculated by the PIV method are shown in Fig.14.

5.2. Measurement of bubbly counter-current flow

In this case, liquid flows downward whereas bubbles rise upward (Fig.15). Two ultrasonic frequencies were used for the measurement. An 8MHz frequency (beam diameter 3mm) was used for liquid velocity measurement. 2 MHz frequency (beam diameter 10mm) was used for bubble measurement. The arrangement of the multiwave TDX and the two-phase flow in the pipe is shown in Fig.15. A typical bubble image of the flow which has been measured is presented in Fig.16. The image was captured

| Pipe I.D. Re number | Pipe 26 mm l.D. | Pipe 50 mm I.D. |
|--|--|---|
| Re = 1,400 (fully developed laminar flow) | L/D = 53 Measured results compared with analytical solution of parabolic velocity profile | |
| Re = 8,100 (fully developed turbulent flow) | | L/D = 46 Measured results verified using PIV data |

Table 2. Measurement conditions for single-phase flow

Table 3. Measurement conditions for bubbly counter-current two-phase flow

| Liquid superficial velocity | 129mm/s |
|---|---------------------------------------|
| Gas superficial velocity | 0.68mm/s |
| Liquid inlet length/D; Gas inlet length/D | 46; 15 |
| Average bubble size | 4 - 5mm |
| Results | Measured data confirmed by PIV method |



Fig.12. Arrangement of the ultrasonic TDX and pipe flow under measurement

using a high speed camera (JVC Hybrid Camera GC-PX1, JVC Co. Ltd.) with frame rate 300Hz.

In this case, measurements have been carried out using both the UVP method (for both phases at the same time) and the PIV method (for the gas phase only, i.e. without seeding particles). Using the multi-wave UVP method, simultaneous measurements of both liquid velocity and gas velocity have been conducted. Instantaneous velocity profiles of both liquid phase and gas phase have been obtained. Using instantaneous values, average velocity profiles of separated liquid phase and gas phase are shown in Fig.17a. Measured results of the UVP method and the PIV method for the gas phase are shown in Fig.17b.

5.3. Accurate measurement of liquid flow rate, deposition, crystalisation and corrosion etc. in piping systems

In industrial piping systems, it is usually desirable to

have highly accurate methods for flow rate measurement at various flow regimes (stationary state, transition state, non-fully developed etc.). Conventional measurement methods are either intrusive or need some assumption of the shape of velocity distribution, e.g. stationary and axisymmetric velocity profile etc. As a result, when the flow regime changes, the accuracy of the measurement cannot be confirmed. Using the measured velocity of the liquid phase in either single-phase flow or two-phase flow, it is possible to calculate the liquid flow rate with high accuracy by integration of liquid flux across the section of the pipe (Fig.18).

Using a measured velocity profile, the flow rate can be calculated using a simple equation $Q = \sum_{i=1}^{n} S_i V_i$ where S_n is the area of the measurement channel on the crosssection of the pipe; V_{n} is the time averaged velocity at the measurement channel. Using this method, there is no need of any assumptions concerning the shape of the velocity profile. Moreover, when more than one TDX are used for the measurement of the velocity profiles at a cross section, the effect of non-axisymmetric velocity distribution can be minimised. This is very helpful in measurement of the flow rate in piping systems with limited length of pipes. In such cases, junctions and bendings will make flows in pipes of short length highly unstable. Velocity profiles are highly nonaxisymmetric. Other measurement methods of liquid flow rate will seriously suffer from such effects.

In addition, using the ultrasonic velocity profile method, measurement of deposition on the pipe wall is enabled. The echo signal captured during measurement clearly exhibits the part of the immobile pipe wall and the part of flowing liquid. Hence, the change in deposition on the pipe wall is monitored and measured directly. At the same time, corrosion of the pipe wall is measured. On the other hand,



Fig.13. Measurement of single-phase flow: (a) fully developed laminar flow (Re=1400); (b) fully developed turbulent flow (Re = 8100)





Fig.14. A typical particle image of single-phase flow (a) and 2D velocity field calculated by PIV method (b)



(a)

5.75 mm

Fig.15. Arrangement of the multi-wave TDX and the two-phase flow in the pipe

Fig. 16. A typical bubble image by high speed camera



Fig. 17. Measurement of two-phase flow: (a) liquid and gas velocity profile measured by UVP method; (b) gas velocity profile measured by UVP and PIV methods



the intensity of echo sound is a function of the purity of the working liquid. Based on this idea, crystallisation of materials in flowing liquid can be measured.

6. Conclusions

The multi-wave UVP and PIV methods for measurement of singlephase and two-phase flow in a vertical pipe have been developed. Measured results using the multi-wave UVP method have been con-

Fig. 18. Velocity distribution in pipe flow

firmed by the optical image processing method (PIV). It has been shown that this method has many advantages and robustness for single-phase and two-phase flow measurement.

Instantaneous velocity profiles of both single-phase and two-phase flows in a vertical pipe have been measured using the multi-wave UVP method. Measured results of single-phase flow agree well with mean velocity profiles of pipe flow. Based on measured velocity profiles of liquid phase and gas phase, other flow parameters can be derived. In addition, using instantaneous velocity profiles of the liquid phase, it is possible to calculate liquid flow rate with high accuracy.

Further research on various pipe flows in the oil and gas industry and in the energy industry using ultrasound will be carried out.

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STUDIES ON TRANSFORMATION OF CO₂ AND CO₂-RICH NATURAL GAS INTO FUELS AND CHEMICALS AT PVPro/VPI: INITIAL RESULTS AND FUTURE WORKS

Dang Thanh Tung, Le Phuc Nguyen Nguyen Hoai Thu, Nguyen Anh Duc Vietnam Petroleum Institute

Summary

This paper summarises some initial research results of CO_2 and CO_2 -rich natural gas transformation into fuels and chemicals at VPI. Our results showed that CO_2 conversion by either direct or indirect routes to fuel or chemical is feasible. The improved catalyst performance by optimisation of catalyst formulation, structure, and catalyst preparation methods will greatly contribute to the overall economics of the process. On the other hand, application of new catalytic reactor technology such as membrane reactor also enhances the process efficiency. These promising results will pave the way for further investment of research activities, aiming to develop new catalysts and technologies that ultimately could help to efficiently utilise CO_2 and CO_2 -rich natural gas in Viet Nam.

I. Introduction

It is believed that natural gas is the most important energy source for the future where natural gas should gradually take over from oil and become the most important fossil fuel by the middle of the 21st century [1]. The abundance of natural gas coupled with its environmental soundness and multiple applications across all sectors means that natural gas will continue to play an increasingly important role in meeting demand for energy as well as being a feedstock for the chemical industry.

Proven natural gas reserves had reached about 196 trillion standard m³ in 2011 [2] without counting the enormous methane reserves trapped in the form of hydrates at the bottom of the oceans. Currently, the majority of natural gas is used for power generation, however, in the longer term it will be increasingly used to produce liquid motor fuels and intermediates for the chemical industry.

Most of the world's natural gas reserves contain some amount of CO_2 , which could be as high as 70 - 80% in volume. With current technology, natural gas is economically viable only when the CO_2 content is less than 10 - 15% of the total volume. If the CO_2 content is higher, then the removal process is prohibitively expensive, therefore leaving these natural gas reserves uneconomical to develop. To be marketable and pipeline ready, the CO_2 content must be reduced at the wellhead to a content level of approximately 2.5%.

Unfortunately, most of the CO_2 that is removed at the wellhead by the gas producers is simply released into the atmosphere. According to the U.S. Energy Information Agency, the production of natural gas is the second largest source of CO_2 emissions in the country. These conditions create a dual economic challenge for the natural gas industry. First, how can we monetise the vast amounts of high CO_2 content as a potential feedstock to produce fuel or chemicals? Second, how can all resource holders, irrespective of the level of CO_2 concentration in their gas, deal with the inevitable need to discontinue the practice of emitting CO_2 during processing?





Vietnam has a total natural gas reserve of approximately 228 billion m³, which mostly locate in the South and central areas, with the CH_4 and CO_2 content in the range of 58 - 81% and 0.1 - 30% respectively. The production capacity is about 8.24 billion m³/year by 2012, which is expected to increase to 13.52 billion m³/year in 2020. Fig.1 shows the balance of gas demand-supply in the South of Vietnam, indicating that there is always a gas shortage in the region [3].

On the other hand, the expected natural gas supply capacity of the central part of Vietnam is 2 - 4 billion m³/ year from 2018 of which 1.3 billion m³/year is reserved for power generation making 0.7 - 2.7 billion m³/year of natural gas available for processing. However, the CO₂ content of the natural gas in this area could be as high as 30% in volume, which imposes some technical difficulties in processing. Furthermore, effective utilisation of CO₂ in some natural gas field could improve the economics as well as environmental protection of the natural gas production in Vietnam. Therefore, the two above-mentioned questions are also important issues for the natural gas industry in Vietnam.

2. PVPro/VPI's initial results

There are two ways to convert CO_2 and CO_2 -rich natural gas to useful fuels or chemical. One is direct conversion of CO_2 into fuels and chemicals, and the other is conversion of CO_2 -rich natural gas into syngas, using CO_2 as a carbonaceous feedstock, followed by other chemical synthesis processes to produce fuels and chemicals. At PVPro/VPI, both of these approaches are adopted to solve challenges in effective utilisation of CO_2 and CO_2 -rich natural gas. The following sections described the initial results of our research projects in these areas.

2.1. Direct conversion of CO, to methanol

In order to directly convert CO_2 to methanol, the CuO/ZnO/Al₂O₃ catalyst system was studied. The objectives are to achieve high yield and selectivity of methanol and to suppress the formation of H₂O and higher

alcohols by the reverse water gas shift (RWGS) reaction to improve the efficiency of the process as well as the catalyst stability. For that, a new procedure for CuO/ZnO/ Al_2O_3 catalyst preparation and modification by CeO₂ addition was studied. Furthermore, the performance of membrane reactor was compared with traditional reactor in CO₂ hydrogenation into methanol to evaluate the possibility to improve CO₂ conversion and methanol yield by using catalytic membrane reactor technology.

2.1.1. New procedure for preparing highly effective catalysts

CuO-ZnO/Al₂O₃ catalysts prepared by co-precipitation method using nitrates of Cu, Zn, and Al have been widely used in methanol synthesis with H_2/CO_2 [4]. In our first study, 30%CuO - 30%ZnO 40%Al₂O₃ model catalysts were prepared by parallel-slurry-mixing method based on [5, 6] with some modifications in 3 different procedures.

Differences of these procedures were the order of mixing γ -Al₂O₃ (prepared in-house) with precipitation of Cu²⁺-Zn²⁺ and use of different precipitation agents. The catalyst prepared by procedure 1 (P1) has the longest contact time between Cu and Zn phases and procedure 2 (P2) ensures the maximum deposition of Cu and Zn on Al₂O₃. Procedure 3 (P3) is combination of the others. The catalysts were symbolised as CZ/Al P1, CZ/Al P2, CZ/Al P3.



Fig.2. TPR profiles of CZ/Al, Cu/Al, 20Ce + CZ/Al, 16Ce + CZ/Al and 8Ce + CZ/Al catalysts

| Table 1. Methanol yield and product selectivity (250°C; 5 bar; 0.5g, GHSV = 36,000h ⁻¹), d_{CHO} and S _{BET} of the ca | talyst |
|--|--------|
|--|--------|

| Catalysts | МТҮ (m _{снзон} (g).Кg _{cat} - ⁻¹ .h ⁻¹) | % CH ₃ OH | % DME (dimethyl ether) | % CO | dCuO (nm) | S _{BET} (m²/g) |
|-----------|---|----------------------|---------------------------|------|-----------|-------------------------|
| CZ/AI P1 | 9.1 | 9.4 | 4.0 | 86.6 | 21 | 70 |
| CZ/AI P2 | 14.4 | 11.1 | 0.0 | 88.9 | 12 | 65 |
| CZ/AI P3 | 25.9 | 12.5 | 0.0 | 87.5 | 27 | 66 |

| Catalysts | MTY (m _{cH30H} (g).Kg _{cat} -1.h ⁻¹) | CH ₃ OH selectivity (%) | CO selectivity (%) | d _{cuo} (nm) | S _{BET} (m²/g) |
|---------------|---|---------------------------------------|-----------------------|-----------------------|-------------------------|
| CZ/AI | 25.9 | 12.5 | 87.5 | 27 | 66 |
| 8%Ce + CZ/Al | 75.6 | 37.5 | 62.5 | 25 | 72 |
| 12%Ce + CZ/Al | 91.2 | 40.7 | 59.3 | 24 | 75 |
| 16%Ce + CZ/Al | 105.4 | 43.9 | 56.1 | 27 | 74 |
| 20%Ce + CZ/Al | 57.4 | 47.4 | 62.6 | 37 | 70 |

Table 2. Methanol yield and product selectivity (250°C; 5 bar; 0.5g, GHSV = 36,000h⁻¹), d_{CuO} and S_{BET} of the catalysts

Table 3. The activity (MTY) of the catalysts for methanol synthesis

| Catalyst | (т _{снзон} (| MTY g).Kg _{cat} ⁻¹ .h ⁻¹) | Reaction conditions | Ref. |
|--|-------------------------------------|--|--|------------|
| | Initial activity Activity after 40h | | | |
| 16%Ce + CZ/Al powder | 105 | 104 | H ₂ /CO ₂ 3/1, 250°C; | This study |
| 16%Ce + CZ/Al tablet | 101 | 98 | 5 bar; 36,000h ⁻¹ | This study |
| Cu/ZnO/ZrO ₂ /Al ₂ O ₃ /Ga ₂ O ₃ powder | 785 | - | H ₂ /CO ₂ 3/1, 250°C; 50 bar; 36,000h ⁻ | [7] |
| Cu-Zn-Ga/SiO ₂ powder | 329 | 349 | H ₂ /CO ₂ 3/1, 250 - 270°C; 20 bar; 36,000h ⁻¹ | [8] |
| Commercial catalyst (Cu/ZnO/Al ₂ O ₃) tablet | 626 | - | H ₂ /CO ₂ 3/1, 250°C; 50 bar; 18,000h ⁻¹ | [9] |
| Cu/ZnO/Ga ₂ O ₃ powder | 738 | - | H ₂ /CO ₂ 3/1, 250°C; 50 bar; 36,000h ⁻¹ | [10] |
| ZrO ₂ -doped CuZnO powder | 780 | - | H ₂ /CO = 2; 210 - 280°C, 50 bar, 4,000h ⁻¹ | [11] |

The results in Table 1 show that catalysts with the same compositions but prepared by different methods have a large difference in their activities. The catalyst prepared by our procedure (P3) has its activity improved by a factor of 1.7 - 3 compared to the others, indicating the importance of not only the catalyst formulation but also the catalyst preparation procedure.

2.1.2. CuO-ZnO/Al₂O₃ catalysts promoted by CeO₂

Table 2 shows that Ce addition could enhance the dispersion of active phase Cu without decreasing the S_{BET} Fig.2 shows TPR-H₂ profiles of catalysts, indicating that Ce addition could increase the reducibility of CuO. By the addition of 8 - 20%CeO₂, catalytic activities of CeO₂-CuO-ZnO/Al₂O₃ catalysts are increased by a factor of 1.6 - 3.0 compared to that of the sample without modification. The highest activity can be obtained with the catalyst having CeO₂ loading of 16wt.%. The decrease of catalytic activity with higher CeO₂ loadings could be caused by the increase of d_{CuO}.

2.1.3. Catalyst shaping and stability test

16%Ce + CZ/Al catalyst formulation was chosen to



Fig.3. Methanol yield versus temperature for both membrane and traditional reactors;

pelletise for industrial application. After the powder has been ground to be suitable for feeding to a pilling machine, it is mixed with a cellulose-typed binder, and then formed into pellets and treated at 280°C. It has been found that treated catalyst pellets still maintain a high BET surface $(60m^2/g)$ and high activity (MTY = 98). Comparisons of CH₃OH productivity of our prepared catalysts with several previous works are given in Table 3. It can be seen that our Ce-based catalyst exhibits a very high stability with time on stream (after 40h on stream).

2.1.4. Application of a membrane reactor to improve the methanol synthesis

The experimental performance of a zeolite membrane reactor has been analysed in comparison to a conventional reaction system. The methanol synthesis from CO_2 and H_2 has been considered versus temperature (Fig.3). The results of this work show a better performance of membrane reactor compared with traditional reactor (TR): at the same temperature, CO_2 conversion for MR was higher than the one related to TR, especially at 220 - 240°C. This improvement can be also seen in the sense that the same methanol yield of TR (at 250°C) can be reached by working with MR over less drastic conditions (at 220°C). This should notably reduce the energy demand.

2.2. CO, reforming of natural gas

In this direction, the harmful greenhouse gases, namely CH_4 and CO_2 , are reformed to produce valuable synthesis gas (syngas). The catalyst based on Ni/Al₂O₃ was developed with some modification to minimise coke formation on the catalyst surface. Our previous study showed that at the optimal active phase concentration of 15wt.%, the conversions of CO_2 and CH_4 were the highest [12]. Therefore, this catalyst formulation was used for our further modification in order to improve catalyst performance.

For that, magnesium oxide was incorporated in γ -Al₂O₃ to enhance the basicity of support because basicity was believed to promote the activation of acidic CO₂

and suppress carbon formation [13]. Cobalt was used as a secondary active component since a small Ni substitution of Co dramatically enhances the catalyst activity and stability by improved resistance to metal oxidation [14, 15]. All catalysts were prepared by co-precipitation method to achieve highly dispersed metallic particles with small crystal sizes.

Catalytic performance of all samples is indicated in Table 4. The addition of MgO could dismiss coke formation on the catalyst surface. The coke deposited on Ni/Al₂O₃-MgO sample was a haft compared to that on Ni/Al₂O₂ sample. The resistance to coking of Ni/MgO-Al₂O₃ catalyst might be assigned for its high sinteringresistance ability and low acidity of support. The results showed that when the cobalt is incorporated in the catalyst, coke formation decreases. The interaction between metallic species Ni° - Co° could cause a synergetic effect that contributes to stabilisation of the catalyst and suppressing coke formation [16]. However, the optimal concentration of cobalt should be about 7.5%. At the Co content of 9wt.%, coke formation on this sample was even higher than that on the sample without Co addition. It could be interpreted by the poorer dispersion of Co on the sample and also the large crystallite size of this sample as shown in Table 4. Our in-house prepared catalyst also showed better performance to a commercial catalyst in term of CH₄, CO₂ conversions as well as H₂, CO yields. The low H₂/CO ratio of the syngas product is also suitable for other downstream processes such as methanol or Fischer-Tropsch synthesis.

3. Future works

Our initial results showed that CO₂ conversion by either direct or indirect routes to fuel or chemical is feasible. The improved catalyst performance by optimisation of

| Catalysts | S _{B.E.T} (m²/g) | d _{ni} (nm) | CH₄ conversion (%) | CO ₂ conversion (%) | CO yield (%) | H ₂ yield (%) | H ₂ /CO | Coke (%) |
|--|------------------------------|----------------------|--------------------------|--------------------------------------|-----------------|-----------------------------|--------------------|-------------|
| 15Ni/ Al ₂ O ₃ | 99 | 5.0 | 73.6 | 73.9 | 50.3 | 51.1 | 1.2 | 20.5 |
| 15Ni / Al ₂ O ₃ - MgO | 82 | 7.0 | 66 | 64.2 | 39.9 | 45.4 | 1.3 | 11.3 |
| 9Ni 6Co/ Al ₂ O ₃ - MgO | 75 | 8.5 | 68.9 | 61.0 | 45.9 | 53.3 | 1.4 | 9.8 |
| 7.5Ni 7.5Co/ Al ₂ O ₃ - MgO | 76 | 10.3 | 65.2 | 62.8 | 46.5 | 53.2 | 1.3 | 9.68 |
| 6Ni 9Co/ Al ₂ O ₃ - MgO | 71 | 11.5 | 65.7 | 63.4 | 40.6 | 51.0 | 1.5 | 13.4 |
| Commercial (Ni/Al ₂ O ₃ -MgO) catalyst | 47 | - | 37.7 | 63.4 | 42.2 | 32.6 | 0.9 | - |

Table 4. Catalytic performance of the catalyst samples ($T = 700^{\circ}$ C, $CH_{4}/CO_{2} = 1.5/1$, GHSV = 12,000ml.h⁻¹.g⁻¹)

catalyst formulation, structure, and catalyst preparation methods will greatly contribute to the overall economics of the process. These promising results pave the way for further investment of research activities, aiming to develop new catalysts and technologies that ultimately could help to efficiently utilise CO_2 and CO_2 -rich natural gas in Vietnam. Obviously, there are still many issues to be solved technically and economically. In order to tackle such challenges, PVPro/VPI has established a long-term R&D programme for deep processing of natural gas, in which the issues of CO_2 and CO_2 -rich natural gas utilisation are of major importance. The objectives of this programme are to be able to identify the suitable technology as well as to develop pilot plants to convert CO_2 or CO_2 -rich natural gas to methanol or fuels with the capacity of 10l/h by 2020.

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PERSPECTIVE USING OF PHENOLIC COMPOUNDS WITH TRICYCLIC ADAMANTYL SUBSTITUENTS AS POLYMERISATION INHIBITORS IN THE PROCESSING OF LIQUID PYROLYSIS PRODUCTS

Do Chiem Tai¹, Dam Thi Thanh Hai¹, W.A.Sokolenko², A.F.Gogatov³ ¹Petrovietnam University ²Institute of Chemistry and Chemical Technology, Russian Academy of Sciences

³A.E Favorsky Irkutsk Institute of Chemistry, Russian Academy of Sciences

Summary

Experimental results demonstrated that two phenolic compounds - 2,6-di-adamantyl-4-methylphenol and 2,4-di-adamantyl-6-methylphenol - are highly effective polymerisation inhibitors in the processing of liquid pyrolysis products (pyrocondensate). In laboratory conditions the two phenols showed very high inhibitory effectiveness in depressing the formation of polymer deposits which could be yielded from unsaturated components of pyrocondensate.

At concentrations of 0.01 - 0.05ppmw the inhibitory effect of 2,6-di-adamantyl-4-methylphenol and 2,4-di-adamantyl-6-methylphenol varied in the interval of 54 - 77% and 36 - 59%, respectively. In comparison to the effectiveness of industrial inhibitor 2,6-di-tertbutyl-4-methylphenol the figures of 2,6-di-adamantyl-4-methylphenol are about 1.7 times higher.

1. Introduction

Angarsk Polymer Plant (in Angarsk City, Russia Federation) provides plastic products and a wide range of monomer, such as ethylene, propylene for producing resins to the Russian market. Through around 40 years of its history, Angarsk Polymer Plant (APP) has always encountered one serious problem. In pyrocondensate processing, under high temperature olefins are usually polymerised and form polymer deposits which fall on the surface of pipeline equipment and decrease heat transfer, hammer plates of distillation columns and lessen the economic effectiveness of petrochemical plants [1].

In APP, practice has shown that the using of polymerisation inhibitors is the most selective and cost-effective

way to deal with the undesirable formation of polymer deposits in pyrocondensate processing, in production, in storage and in transportation of monomers and fuels. The main role of inhibitors is to depress or/and reduce the rate of radical reaction of unsaturated compounds which usually occurs under high temperature. The assortment of compounds used as such specific additives is very wide. Because of their efficiency and adaptability, phenolic compounds are the most common polymerisation inhibitors used in processing unsaturated components of liquid pyrolysis products, especially in the Russian petrochemical plants [2].

Experience of APP's experts in using inhibitors over 30 years showed that the most widely-used and potent polymerisation inhibitors are sterically hindered phenols due to their high efficiency, low toxicity, high thermal stability and high antioxidant activity [2 - 4].

According to [5] phenols with the hydroxyl group shielded by large substituents have high solubility in organic solution and high anti-radical activity. For example, the above-mentioned properties of isobornylphenols (organic compounds with bicycle substituents) have been



Fig.1. The surface of fractionator's trays in the case of using highly effective phenolic inhibitor and uneffective nitroxyl radicals

confirmed by the results of laboratory experiments in pyrocondensate processing [6] and also by the data of antiradical activity in reaction with stable 2,2-diphenyl-1-picrylhydrazyl radical (DPPH) [7].

The experimental data [7] showed that among isobornylphenols only compounds whose hydroxyl group on both sides is shielded by isobornyl substituents have the highest anti-radical and inhibitory activity. Furthermore, these properties depend significantly on the structure of phenols: on the number of large substituents and on the mutual positions of substituents in the molecule relative to the reactive centre (OH-group). So it is expected that phenolic compounds with larger (tricyclic) substituents could have higher inhibitory effectiveness. From this point, phenolic compounds with adamatyl substituents should be perspective candidates. It is important to be reminded that some good properties of adamatyl phenols, such as biological activity, thermal stability, and low toxicity, have already been shown in some papers [13, 14]. However, the activity of these substances in inhibiting polymerisation reactions which usually occur under high temperatures in the processing of liquid pyrolysis products has not yet been studied.

In addition, it is well known that adamantane and its derivatives can be obtained at the petrochemical plants from its own materials - from cyclopentadiene-dicyclopentadiene (CPD-DCPD) fraction - by simple catalytic reactions [8]. For example, the industrial method for synthesis of adamantane from CPD has been studied and well organised (Fig.3) [9]. Various strong Lewis acids, such as AlCl₃ and SbCl₃ can be used as catalysts for this reaction. Moreover, the synthesis of adamantylphenols has now been fairly well mastered [10 - 12].

From the foregoing, the testing of inhibitory activity of adamantylphenols in pyrocondensate processing is essential and shows a certain scientific interest.

2. Experiment

2.1. Purpose of study

Evaluating the effectiveness of monohydroxy adamantylphenols in inhibiting formation of polymer deposits which occurs in pyrocondensate under high temperature; the experiment was carried out in a laboratory condition $(130 \pm 2^{\circ}C)$ which was similar to the production condition of the Angarsk Polymer Plant (APP).



Fig.2. Antiradical activity of isobornylphenols defined by the reaction with DPPH [7]



Fig.3. Synthesis scheme of adamantane from cyclopentadiene

 Table 1. Essential unsaturated compounds of pyrocondensate K-20, wt. %

| Components | Quantity, % mass | | | |
|-----------------------------|------------------|--|--|--|
| 2-methyl-butene-1 | 1.53 | | | |
| 3-methyl-butene-1 | 0.32 | | | |
| 2-methyl-butene-2 | 0.77 | | | |
| 1-butyne | 0.37 | | | |
| 1,2-butadiene | 0.12 | | | |
| 1,3-trans-pentadiene | 2.10 | | | |
| 1,3- <i>cis</i> -pentadiene | 1.67 | | | |
| 1,4-trans-pentadiene | 0.75 | | | |
| 1-pentene | 1.09 | | | |
| 3-ethyl-pentene-1 | 6.45 | | | |
| 4-methyl-pentene-2 | 0.46 | | | |
| cis-pentene-2 | 0.32 | | | |
| trans-pentene-2 | 0.60 | | | |
| isoprene | 1.18 | | | |
| cyclopentene | 1.37 | | | |
| 3-ethyl-pentene-1 | 6.45 | | | |
| styrene | 0.31 | | | |

Table 2. Distillation fractions of pyrocondensate which were determined on "АРН-ЛАБ-03"

| Temperature (°C) rea | Temperature (°C) reaching after different amount (% vol.) of pyrocondensate K-20 was distillated | | | | | | | | | |
|------------------------|--|------|------|------|------|------|------|------|-------|-------|
| Starting boiling point | 10 | 20 | 30 | 40 | 50 | 60 | 70 | 80 | 90 | 97.5 |
| 44.0 | 56.0 | 60.5 | 66.0 | 70.5 | 74.0 | 81.0 | 85.5 | 97.5 | 147.5 | 173.0 |

2.2. Subjects of study

In this research, liquid hydrocarbon mixtures of fractionator N°-20 of the technological pipeline of APP (abbreviated as pyrocondensate K-20) were used as the objects for evaluating the inhibitory effectiveness of 2,6-di-adamantyl-4-methylphenol and 2,4-di-adamantyl-6-methylphenol.

Before each series of experiments the composition of pyrocondensate K-20 was studied by the Scientific and Research Centre (S&RC) of APP by chromatography mass spectrometry "Hewlett Packard-5MS" using the database of Angarsk Petrochemical Complex. The results are presented in Table 1.

The fractions of pyrocondensate K-20 were determined on the "APH-ЛАБ-03" using the method of determining the fractions of the petroleum products (Table 2). In addition, some other properties of pyrocondensate K-20, provided by S&RC, are showed in Table 3.

2.3. Adamantylphenols used in this research

Phenolic compounds - adamantylphenols - used in this program were synthesised and provided by W.A. Sokolenko - senior researcher of the Institute of Chemistry and Chemical Technology, Siberian Branch of the Russian Academy of Sciences. These compounds are:

2,6-di-(1-adamantyl)-4-methylphenol (26DA4MP) (Fig.4a): synthesised by alkylation of p-cresol with 1-adamantanol at temperature 18 - 20°C in the presence of trifluoroacetic acid (TFA) as a catalyst [15]. 26DA4MP is white crystalline substances melting at 262 - 263°C.

2,4-di-(1-adamantyl)-6-methylphenol (24DA6MP) (Fig.4b): obtained by the reaction between o-cresol and 1-adamantanol in TFA catalyst [16]. This organic compound is a white crystal whose melting point is 245 - 247°C.

2.4. Methods of studying the inhibitory effectiveness of adamantylphenols

2.4.1. Evaluating the inhibitory effect of adamantylphenols using Budarov's method

By this method, the inhibitory effective-

ness of adamantylphenols was determined by metering the inhibitor into an autoclave with pyrocondensate. Then, the autoclave with pyrocondensate and a certain amount of inhibitor experienced heat treatment at 130°C for 1 hour. Thence, the mixture was cooled to room temperature (~22°C), and the determination of polymer deposits formed after heating was carried out in appliance with the Russian GOST 8489-85 [17] on special device "ПОС-77M" (Fig.6).

A typical experiment includes the following steps:

- 100ml pyrocondensate was poured into a stainless steel autoclave (a type of reactor working under high pressure), and then a certain amount of inhibitor in the range from 0.01 - 0.05wt. % in comparison to mass of pyrocondensate was added to the autoclave.

- Then, the autoclave was carefully sealed and put into an oil thermostat which was already preheated to 130 \pm 2°C. The temperature was kept at this level for 1 hour, then the autoclave was taken out and cooled down to room temperature;

Table 3. Other properties of pyrocondensate

| Properties | Pyrocondensate K-20 |
|--|----------------------------------|
| lodine number, g l ₂ /100g pyrocondensate | 92 |
| Average molecular weight, g/mole | 87 |
| Fractional composition | Primarily fraction C_5 - C_7 |



Fig.4. Structure of 2,6-di-(1-adamantyl)-4-methylphenol (a) and 2,4-di-(1adamantyl)-6-methylphenol (b)



26DA4MP: $R_1 = R_3 = R_6 = H$; $R_2 = R_6 = CH_3$; $R_4 = R_7 = Ad$ 24DA6MP: $R_1 = R_2 = R_5 = H$; $R_3 = R_7 = CH_3$; $R_4 = R_6 = Ad$



Fig.5. Synthesis scheme of 26DA4MP and 24DA6MP



Fig.6. Special device "ΠΟC-77M" used for determination of polymer deposits

- In the next stage, pyrocondensate was distilled by water vapour at 160°C on the device type "ΠOC-77M" and the polymer deposits were weighed (C,);

- A parallel experiment without inhibitor was carried out, and then the quantity of polymer deposits was measured (C_{0});

- Inhibitory effect of the inhibitor was determined by correlating the mass of the polymer deposits formed in the presence of the inhibitor with the mass of polymer deposits formed in the absence of the inhibitor. Inhibitory effect was calculated by the following formula:

$$E(\%) = \frac{C_o - C_i}{C_o} \times 100$$
 (1)

The discrepancies between the results of two parallel experiments should not exceed the values given in Table 4.

 Table 4. Acceptable discrepancies between the results
 of two determinations

| Amount of polymer deposits, mg/100cm ³ of pyrocondensate | Permissible differences |
|---|-------------------------|
| Up to 15 | 2mg |
| Above 15 to 40 | 3mg |
| Above 40 to 100 | 8% of the lower result |
| Above 100 | 15% of the lower result |

2.4.2. Determining inhibitory effect of adamantylphenols using the iodine number

The iodine number of pyrocondensate was determined by a standard method - the Russian GOST 2070-82 "Methods for determining the iodine number and concentration of unsaturated hydrocarbons" [18]. This method is widely used to determine the iodine number and the concentration (mass %) of unsaturated hydrocarbons in gasoline, diesel fuel and other light petroleum products.

The method is carried out by mixing alcoholic solution of iodine and pyrocondensate (gasoline, diesel fuel and other products). After that, the titration of the alcoholic solution with sodium thiosulfate solution ($Na_2S_2O_3$) is undertaken to determine the mass of free iodine (I_2) which did not react with unsaturated compounds in the pyrocondensate:

$$I_2 + 2Na_2S_2O_3 \rightarrow 2NaI + Na_2S_4O_6$$

Then the amount of iodine (g) involved in the reaction with unsaturated compounds containing in 100g pyrocondensate is evaluated.

lodine number (Z) of pyrocondensate is calculated by the following formula:

$$Z = \frac{100 \times (V_1 - V_2) \times 0.012692 \times F}{m}$$
(2)

Where:

 V_1 (cm³): The amount of sodium thiosulfate consumed in the titration of iodine in a blank experiment (without pyrocondensate);

 V_2 (cm³): The amount of sodium thiosulfate consumed in the titration of iodine in the experiment with pyrocondensate;

F = 1: Factor of sodium thiosulfate solution 0.1mol/ dm^3 ;

m: The mass of pyrocondensate;

0.012692: The amount of iodine contained in 1 cm^3 solution of Na₂S₂O₃ 0.1N.

The iodine number of pyrocondensate is the average value of iodine numbers calculated from four parallel experiments, and then the iodine number is rounded to 1 decimal digit.

By the iodine number method, the inhibitory effect of the inhibitor is calculated using the following formula:

$$E(\%) = \frac{(Z_{130,(2)} - Z_{130,(1)})}{(Z_{20} - Z_{130,(1)})} \times 100$$
(3)

Where:

 $Z_{130, (1)}$ and $Z_{130, (2)}$: lodine number of pyrocondensate after heat treatment at 130°C in the blank experiment (without inhibitor) and in the experiment with inhibitor, respectively;

 Z_{20} : lodine number of pyrocondensate without inhibitor before heat treatment (20°C).

3. Experimental results and discussion

The data were determined by a series of experiments in laboratory conditions. The methods for determining inhibitory effects were introduced above (section 2.4).

Phenolic compounds with adamantyl substituents were thoroughly dissolved in pyrocondensate. Therefore, they can be dosed directly without using auxiliary solvents.

The experimental data showed that after heat treatment of pyrocondensate K-20 in the absence of inhibitor the level of polymer deposits was very high and reached 375mg/100cm³ pyrocondensate. The figures decreased strongly to 206 - 262mg in the case of using ionol (Fig.7). It can be easily seen that in the presence of 26DA4MP the number dropped significantly to 85 - 171mg. This indicated that 26DA4MP was more effective than the widely used inhibitor ionol.



Fig.7. Amount of polymer deposits formed in pyrocondensate K-20 at different concentration of 26DA4MP and lonol



Experimental results presented indicate that the compound 26DA4MP had high activity in inhibiting radical reactions of unsaturated components of pyrocondensate K-20. Indeed, on effectiveness, 26DA4MP far exceeds ionol - industrial inhibitor (Fig.8). In the range of inhibitor concentrations from 0.01 - 0.05 wt.% (or ppmw), inhibitory effectiveness of 26DA4MP reached 54 - 77% while the data of ionol gave the values of 30 - 45%. We can see that inhibitory effects of the two inhibitors increased proportional to their concentrations in pyrocondensate, and at any concentration the inhibitory effects of 26DA4MP were superior to the figures for ionol. It should be emphasised that at concentration 0.01wt.% the effectiveness of 26DA4MP reached 54% which was already higher than the data of ionol (45%) achieved at concentration 0.05wt.%.

We considered that the reasons for the higher inhibitory effectiveness of 26DA4MP in comparison to ionol are the following:

- First of all, 26DA4MP had higher solubility in pyrocondensate than ionol. At room temperature (~24°C) the average solubility of 26DA4MP in pyrocondensate is about 93mg/cm³ while the number for ionol is only around 56mg/cm³. Practice showed that if an inhibitor has good solubility in pyrocondensate (or in objects which need to be protected from polymer deposits formation) then the system *"inhibitor + pyrocondensate"* is more homogenous. Consequently, the inhibitory effectiveness of the inhibitor is high because the radical reactions between inhibitors and alkyl radicals usually are homogenous [20, 21].

- It is well-known that in depressing the formation of polymer deposits by using phenolic inhibitors the main process is the reaction of phenols (PhOH) or phenoxyl radicals (PhO•) with alkyl radicals:

 $PhOH + R \bullet \to PhO \bullet + RH \tag{4}$

$$PhO_{\bullet} + R_{\bullet} \rightarrow Ph_{1}O_{\bullet} + R_{\bullet}$$
(5)

Unfortunately, some undesirable secondary reactions, such as reaction O-O-recombination of phenoxyl radicals usually occur parallel with the main reactions.

$$PhO_{\bullet} + PhO_{\bullet} \to Ph-O^{-}O-Ph$$
 (6)

 $PhO_{\bullet} + Ph_{1}O_{\bullet} \rightarrow Ph-O_{\bullet}O_{-}Ph_{1}$ (7)

$$Ph_1O \bullet + Ph_1O \bullet \to Ph_1 - O^-O - Ph_1$$
(8)

However, the rate of reactions (6), (7) and (8) is high only if two ortho- positions from both sides of OH- group in phenols are small substituents. From this point, 26DA4MP was more effective than ionol because adamantyl groups are much larger than tert-butyl groups, so they prevented the contact of two phenoxyl radicals created from 26DA4MP. A possible schematic reaction O-Orecombination of phenoxyl radicals formed from phenolic inhibitors is presented in Fig.9.

From hypothetical schemes it could be understood that in pyrocondensate processing both directions (a) and (b) are not desirable because they lead to inefficient consumption of inhibitors. However, in the case of lonol, direction (a) could occur because two *tert*-butyl substituents are not large enough to prevent the reaction O-O-recombination. On the other hand, reaction O-Orecombination (b) could not occur because two adamantyl chains are very large. Therefore, two phenoxyl radicals created from adamantyl phenol could not approach closely enough and react with each other.

The higher the rate of polymer deposits formation the lower the concentration of unsaturated compounds in pyrocondensate after heat treatment, and vice versa. In addition, the quantitative characteristic of the content of unsaturated hydrocarbons in pyrocondensate can be estimated by the iodine index of pyrocondensate.

It is clear that the higher the content of unsaturated compounds in pyrocondensate, the higher the iodine index of pyrocondensate. Based on this argument, iodine numbers of pyrocondensate before and after heat treatment in the case of using inhibitors and without inhibitors could be used as an independent method to assess inhibitory activity of ionol and 26DA4MP. The iodine values were calculated by formula (2) and presented in Table 5 and Fig.10.

The Fig.10 showed that at the concentrations in the range of 0.01 - 0.05% wt. in the case of using 26DA4MP as an inhibitor, iodine numbers of pyrocondensate reached the level of 63 - 76mg $I_2/100$ mg pyrocondensate. When ionol is used the figures dropped to 50 - 58mg $I_2/100$ mg pyrocondensate. In addition, it is clear that iodine numbers of both inhibitors were proportional to their concentrations in pyrocondensate.

By iodine number, the inhibitory effect can be calcu-



Fig.9. Reaction O-O-recombination between phenoxyl radicals formed from ionol (a) and from 26DA4MP (b)

Table 5. lodine number of pyrocondensate K-20

| | | Temperature, °C | lodine number (Z) of pyrocondensate in case of using inhibitors at different concentration (% wt.) | | | | | |
|-----------|---------|--------------------|--|------|------|------|------|--|
| | | | 0.01 | 0.02 | 0.03 | 0.04 | 0.05 | |
| Inhibitor | WI * | 20** | 92.0 | | | | | |
| | | 130*** | 30.8 | | | | | |
| | lonol | 130*** | 50.4 | 54.1 | 54.3 | 58.5 | 57.9 | |
| | 26DA4MP | 130*** | 62.8 | 68.8 | 70.2 | 76.4 | 77.5 | |

* - WI: lodine number of pyrocondensate without inhibitor; **: lodine number of pyrocondensate before heat treatment; ***: lodine number of pyrocondensate after heat treatment at 130°C

lated by formulation (3). The results are presented in Fig.11. As mentioned above, the higher the iodine index the higher the content of unsaturated compounds in pyrocondensate after heat treating under high temperature ~130°C, and consequently, the higher the inhibitory effect.

Based on inhibitory effectiveness determined by the two independent methods we can see that the inhibitor 26DA4MP was much more effective than industrial lonol. Indeed, the inhibitory effects of 26DA4MP varied in the interval of 60 - 80% and about 1.63 times higher than the figures of ionol.



Fig.10. lodine number of pyrocondensate with inhibitor ionol or 26DA4MP



Fig.11. Inhibitory effect of ionol and 26DA4MP evaluated by iondine number

In comparison to 26DA4MP, its analogue - compound 24DA6MP - showed lower levels of inhibitory effect. However, this compound was still more effective than ionol.

The graph in Fig.12 shows that in the presence of 24DA6MP the quantity of polymer deposits achieved the level of 154 - 241g/100cm³ pyrocondensate K-20. On the other hand, the data rose significantly to 206 - 262g/100cm³ in the case of using ionol as inhibitor. This means that 24DA6MP was more effective in protecting unsaturated components from polymerisation reaction than ionol.

The experimental data (Fig.13) showed that at any concentration in the range of 0.01 - 0.05wt.% 24DA6MP's effects were higher than the figures for the industrial inhibitor - ionol. It was clearly seen that in the interval of 0.01 - 0.03wt.% the data of 24DA6MP increased from 35 - 58%. However, the figures remained stable at this level despite its concentration rising from 0.03 - 0.05wt.%. This indicates that a high concentration of 24DA6MP in pyrocondensate was not needed.



Fig.12. Amount of polymer deposits formed in pyrocondensate K-20 at different concentration of 24DA6MP and lonol



Fig.13. Inhibitory effect of 24DA6MP and ionol

It is supposed that at a high concentration of 24DA6MP, the unwanted O-O-recombination reaction of phenoxyl radicals (Fig.14) occurred more slowly than that presented in Fig.9a as the result of sterical effect of a large adamantyl group. However, the former could occur much faster than the O-O-recombination reaction showed in Fig.9b.

According to this scheme (Fig.14), phenoxyl radicals, which were formed from 24DA6MP, could rotate around its C-O axis and choose the most appropriate position to react with each other. Because of the small size of methyl group phenoxyl radicals formed from 24DA6MP, they were more active in participating in O-O-recombination reaction, and therefore, less effective in inhibiting the formation of polymer deposits.

It is very interesting that the change of relative positions of the methyl and adamantyl groups in the benzene rings (methyl group from para- position to ortho- position and adamantyl group from ortho- position to para- position) led to significant decrease of the inhibitory effect of



Fig. 14. The inefficient consumption of inhibitor 24DA6MP

Table 6. lodine number of pyrocondensate K-20

| | Temperature, | | lodine number (Z) of pyrocondensate in case of using inhibitors at different concentration (% wt) | | | | | | |
|-----------|--------------|--------|--|------|------|------|------|--|--|
| | | C | 0.01 | 0.02 | 0.03 | 0.04 | 0.05 | | |
| Inhibitor | WI * | 20** | 92.0 | | | | | | |
| | | 130*** | 30.8 | | | | | | |
| | lonol | 130*** | 50.4 | 53.4 | 54.3 | 58.0 | 58.3 | | |
| | 24DA6MP | 130*** | 52.6 | 56.0 | 59.7 | 63.3 | 66.7 | | |

*: WI: lodine number of pyrocondensate without inhibitor; **: lodine number of pyrocondensate before heat treatment; ***: lodine number of pyrocondensate after heat treatment at 130°C



Fig.15. lodine number of pyrocondensate with inhibitor ionol or 24DA6MP



Fig.16. Inhibitory effect of ionol and 24DA6MP evaluated by iondine number

phenolic compounds. This demonstrated that the sterical effect of large groups at ortho-positions (of benzene ring) is one of the key factors creating high inhibitory effectiveness of phenolic compounds. We also carried out the determination of the iodine index of pyrocondensate to study the inhibitory effectiveness of 24DA6M. The experimental figures are presented in Table 6 and Fig.15.

According to the experimental data, as the concentration of inhibitors varied from 0.01 - 0.05wt.% iodine numbers of 24DA6MP were higher than the figures of ionol by around 2 - 9 units. It means that 24DA6MP more effectively saved unsaturated components of pyrocondensate from polymeri-

sation reaction than lonol. Indeed, in the concentration range from 0.01 - 0.05wt.% the inhibitory effectiveness of ionol archived 32 - 45% while in this range of concentration the number of 24DA6MP increased from 36 - 59% (Fig.16).

4. Conclusions

The experimental results in laboratory conditions showed that two monohydroxy phenols with adamantly substituents at ortho- positions were highly effective polymerisation inhibitors in pyrocondensate processing. The inhibitory effect of 26DA4MP achieved 57 - 78% and is higher than that of ionol - widely used industrial polymerisation inhibitor - at around 19 - 20%. In addition, the study on the inhibitory effect of the two adamantylphenols has opened a new way of using phenolic compounds with tricyclic substituents as highly effective polymerisation inhibitors. It is considered that the use of adamantylphenols as inhibitors for processing of liquid pyrolysis products of Angarsk Polymer Plant is likely to bring high economic efficiency to the plant in the future. However, some testing of these compounds in an industrial scale trial is needed before using them in the plant's technological pipeline.

With the proven advantages on inhibitory effectiveness and, in addition, relatively simple synthesis and cheap source of raw materials, the use of these substances for petrochemical facilities, for manufacturing monomers, and for stabilising petroleum products in Vietnam is essential. However, in order to accurately assess the economic efficiency, some research on the specific products produced in Vietnam will be needed.

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SOLUTIONS FOR LOCAL ISOLATED GRID WITH HYBRID SYSTEM INCLUDING WIND TURBINE INTERCONNECTION

Nguyen Duc Huy¹, Tran Nam Trung², Tran Khanh Viet Dung³ Nguyen Phung Quang¹, Vo Hong Thai⁴

¹ Hanoi University of Science and Technology

- ² Vietnam Electricity
- ³ Vietnam Oil and Gas Group
- ⁴ Vietnam Petroleum Institute

Summary

The interconnection of distributed energy systems (renewable energies, diesel generators, and hybrid systems etc.) with utility grid has been one of the most important R&D orientations for many years. The majority of the utilities in the world were not conceived to accommodate large-scale distributed energy systems which can cause problems to the utility grid and different solutions have been developed and proposed. However, the hybrid system (wind turbines and diesel generators) with large capacity of wind turbine interconnection to the local isolated electrical grid on an island has not yet gained a comprehensive solution. In this paper, we present the solution for our hybrid system in Phu Quy island - Vietnam, which has 3 wind turbines (3 x 2MW) and 6 diesel units (6 x 0.5MW) interconnected to the local isolated grid (22kV). Our goals are to maximise the penetration of wind power and keep the system stable. Analytical and simulation studies were performed in order to validate the accuracy of the proposed solution. The results were approved by Vietnam Electricity and the solution proposed is currently being used for the stable operation of our hybrid system.

1. Introduction

Phu Quy is the largest island of Cu Lao Thu, in Binh Thuan province, Vietnam. The island's area is approximately 16.52km² with 24,000 residents. The main generation source for the island up until 2012 was 6 diesel generators with total capacity of 3MW, commissioned in March 2009. This source of energy met the basic residential demand and part of industrial usage on the island. The downsides of using diesel generation were high cost (~ 24US cents/ kWh) and high carbon emissions [1]. This imposed a restriction on economic and social development, trading and commercial services on the island. The current load on the island is primarily residential (~ 2MW). Phu Quy island is in the region of the highest wind potential in Vietnam - average wind speed over a year is around 9m/s at 60m hub height. This is an ideal speed for developing wind power. The wind power project in Phu Quy island with the total capacity of 6MW was commenced in 2010 and commissioned in January 2013.

The wind-diesel hybrid system in Phu Quy island currently consists of 6 diesel units with capacity 6 x 0.5MW and 3 wind turbines 3 x 2MW (Vestas V80). While operating this hybrid system, there existed a number of technical challenges that needed resolving including designing an operation scheme for the hybrid system and developing a suitable SCADA system for operational purposes. In addition, research has been undertaken to improve the penetration of wind power in order to reduce the electricity tariff and the amount of carbon released from burning fossil fuel on the island.

Actually, there are a number of applications in the world that currently deploy the wind-diesel hybrid configuration such as the isolated grid in Alaska-US [2], those in remote areas in Australia and larger scale applications in the Canary islands, Spain [3]. The technical constraints for each particular network depend on several factors: wind turbine technology, ratio of installed wind capacity over diesel generation and load characteristics (controllability and reliability required). Therefore, there is no universal solution for wind-diesel hybrid systems; an optimal operation scheme needs to be developed for each case.

The power system in Phu Quy island belongs to the high penetration class according to NREL's classification [2]. During the development and operation of the system, there existed a number of technical challenges that needed resolving. This paper presents results found from a study to improve Phu Quy system's stability, reliability and penetration of wind power. The structure of this paper is as follows: Section II introduces technical constraints for the hybrid system in Phu Quy island, Section III gives an overview and analysis of solutions for enhancing the wind power penetration. Section IV presents a financial analysis for investing in a low-load diesel generator. Finally, conclusion and recommendations are provided in Section V.

2. Current operation scheme and technical costraints

2.1. Constraints on frequency stability

In a 3-phase power system with synchronous machines, keeping the system frequency within acceptable tolerances is equivalent to maintaining a good balance between generation power and demand. In such a system the synchronous generator speed is always kept at the synchronous speed. The rotor inertia, turbine inertia and speed governor play an important role in stabilising and keeping the system frequency at 50/60Hz.

The principle of grid connected synchronous machines does not apply to wind turbine generators. Wind turbines nowadays utilise a doubly-fed induction generator (DFIG) or permanent magnet synchronous generator connected to grid via a bi-directional power electronic converter. The rotor speed can be varied within a



Fig.1. Frequency response to a load switching event with different number of diesel generators



Fig.2. Frequency response to sudden load increase at different penetration levels

wider range in order to harvest maximum wind energy. For these two technologies, the rotor inertia does not play a role in stabilising the system frequency. Therefore, a common problem for a system with a high penetration level of wind power is reduced inertia [3]. When large switching events occur, the system frequency changes affecting equipment operation and may trigger frequency relays.

For the power system in Phu Quy island, due to the high ratio of wind power capacity over diesel generation capacity, the number of diesel generators operating simultaneously plays an important role in the system inertia. The phenomenon of reduced inertia can be clearly seen from a partial load shedding simulation in Fig.1.

On the other hand, simulations also show that with the same number of diesel units operating, increasing the power ratio of wind power has negligible impact on the system frequency response (Fig.2).

The existing Cummins diesel generators in Phu Quy island have reasonably good frequency response and controllability. In practice, with more than 3 diesel generators operating, the hybrid system is relatively stable.

According to technical references, Vestas V80 wind generators also have the capability of adjusting power generated to the system frequency thus helping improve the stability of system frequency. However, this function has not yet been activated.

2.2. Constraints on voltage stability

In addition to the requirement of balancing active power, a power system also requires a balanced reactive power. Balancing reactive power is directly related to the grid voltage. Cummins diesel generators in general have good reactive power generation capability with rated power factor in the range of 0.8 - 0.85.

The capability of generating reactive power of wind turbine generators in Phu Quy island is limited: the generator power factor falls in the range of 0.98 - 1. This means wind generators have little ability in controlling voltage at the connection point. The load in Phu Quy island is mainly residential with very high power factor (0.92 - 0.93). The load therefore consumes very little reactive power and operational experience shows insignificant technical problems relating to voltage stability. However, the limited capability of generating reactive power from wind turbines would become a challenge when increasing the penetration level. When the penetration level increases, the number of diesel generators decreases leading to a drop in reactive power reserve. Besides, the use of diesel generators to meet reactive power demand results in those generators operating at a low power factor meaning low efficiency.

2.3. Constraints on diesel generator operating modes

In addition to maximum power constraints, diesel generators are also restricted by minimum power generated (~ 30% rated power). This is a barrier for increasing wind power penetration. To maintain system stability, there must be a minimum number of diesel generators operating to maintain the system's inertia (2 - 3 generators with existing loading). Therefore the wind power penetration is also limited by the minimum power generated by diesel generators.

2.4. Spinning reserve

Maintaining spinning reserve and responding to load variations is also an important technical issue for the hybrid system in Phu Quy island. At the moment, frequency control is governed by the diesel units. Diesel generators respond in the first place when there are load variations. The SCADA system detects the change in load and adjusts the power generated by wind turbines to ensure the predefined wind penetration. Due to the slow update duration of the SCADA system (in terms of seconds), wind turbines do not play a role in spinning reserve and primary response. This is an enormous barrier when trying to increase the wind penetration. As discussed in Section 2.1, enabling the frequency response



Fig.3. Power constraints of Cummins diesel generators

function of Vestas wind turbines would help wind power in providing primary response.

2.5. Minimum wind power

With wind speed over 7m/s, the minimum wind power generated by V80 generator is 500kW. When the wind speed increases over 17m/s, the minimum wind power generated is 800kW. At a certain load range, these restrictions limit the number of wind turbines operating since if all wind turbines are used this would violate the maximum allowable penetration.

2.6. Current operation scheme

The maximum allowed wind penetration at the moment is 50%. Considering all constraints given above, the operation scheme for the Phu Quy hybrid system is illustrated in Fig.4. This operation scheme has been developed based on the fact that diesel generators are responsible for all spinning reserve of the system ensuring high reliability. Results in Fig.4 show that 4 diesel generators are required at maximum load and 3 diesel generators are required at lower load.

If the wind penetration is not limited, in order to meet all technical constraints discussed above, the operation scheme is illustrated in Fig.5. The maximum wind penetration reaches 69% at peak load.

If comparison is made between the two operation schemes in Figs.4 & 5, it is clear that the number of diesel generators remains unchanged when the penetration level increases. This is due to the spinning reserve requirement. Nonetheless, diesel generators would have to operate at a low power factor in this case (approximately 0.6).

3. Overview of auxiliary technical solutions for the wind-diesel hybrid system

Auxiliary solutions to improve stability and efficiency in hybrid system can be classified into 3 categories as follows: auxiliary equipment to improve stability, low-load diesel generators to improve the penetration level and energy storage devices.

3.1. Auxiliary equipment to improve stability

Auxiliary equipment used for improving stability includes flywheels and dump-loads. Dump-loads have been used widely in wind powered systems in Alaska with the aim of consuming excess energy generated by wind tur-





and satisfying all technical constraints

bines. Dump-loads are generally equipped with a control system enabling fast response. Flywheels developed by Beacon [4] and ABB [5] consist of a rotating element which stores rotational dynamic energy and a bi-directional converter which allows either storing or releasing the energy of the flywheel. Advantages of a flywheel include very fast response (milliseconds) and high efficiency. However, the downside of the device is its limited storage capacity (order of kWhs) and it can only be used to adjust the frequency in a short duration.

The use of a flywheel does not increase the penetration level of wind power, it only improves the system stability and reliability. It is therefore difficult to quantify the financial benefits from a flywheel. On the other hand, the use of a dump-load may resolve the issue of minimum wind power (section 2.5), enabling higher penetration level at certain loading levels. As a consequence, a dump-load investment can be considered on the basis of having a better possibility to improve the wind power penetration.

3.2. Low-load diesel generator

Unlike conventional diesel generators which have to operate at a minimum 30% rated power, low-load diesel

generators are particularly designed to operate efficiently at 5% loading. This helps to reduce diesel operating costs, and at the same time utilising generators to govern the frequency, voltage and spinning reserve. The solution of using a low-load diesel generator is particularly useful for isolated hybrid systems of wind-diesel. This solution has been applied at several isolated systems such as Coral Bay (900kW wind power + 2240kW diesel), Delham (920kW wind power + 1,920kW diesel). The financial model for analysing an investment of a low-load diesel generator is relatively simple with low uncertainties.

3.3. Auxiliary energy storage devices

Energy storage devices include pumped-storage, fuel cells, batteries of different technologies integrated with power electronic converters [6]. One of many advantages of energy storage devices is the fact that almost all of wind energy can be exploited. Storage devices will store energy when wind power is larger than load demand and will release the energy when wind power ceases. In addition to their storage function, such devices can be integrated with control functionality to stabilise frequency, voltage and spinning reserve.

Each energy storage device has its own technical characteristics and features. Fuel cells normally have large capacity and energy. Super capacitors have good efficiency and fast response, however the amount of stored energy is limited. A common feature of storage devices is their high cost therefore an investment has to be thoroughly considered.

The possibility of using storage devices to store energy and make the most of wind energy depends on the correlation between wind power and load demand. With the present condition of load demand in Phu Quy island, the use of storage devices is not feasible because with 3 wind turbines of 2MW each, at all times, dispatchable power is around 2 - 2.1MW (30% of installed capacity). This power level is still higher than peak load in Phu Quy island, not to mention, the wind power needs to be limited due to the requirement of diesel operation to control the frequency and voltage. Preliminary calculations show that it would be inefficient to invest in a storage device on Phu Quy island in the near future.

Table 1 presents a qualitative comparison between different auxiliary solutions which can be applied to the Phu Quy power system. It is obvious that the low-load diesel generator solution meets reasonably well all technical requirements with a simple operation scheme. Hence this solution should be carefully considered. Besides, the solution of using a compensation capacitor in combination with a dump-load also proves useful as it allows improving stability and penetration level and at relatively low costs.

4. Financial analysis for investment in a low-load diesel generator

Table 1. Comparison of auxiliary solutions

| Auxiliary device | Frequency stabilisatio | Spinning reserve | Voltage Stability | Enhance penetration | Costs |
|-----------------------|------------------------|---------------------|----------------------|---------------------|-------|
| Flywheel | +++ | - | + | - | High |
| Dump-load | + | - | - | + | |
| Fuel cell | ++ | ++ | ++ | ++ | |
| Super Capacitor | +++ | + | +++ | + | High |
| LL diesel | ++ | +++ | ++ | +++ | |
| Capacitor + Dump load | + | - | + | + | Low |

A computational model for analysing optimal investment for an auxiliary solution is a complex one as it depends on a number of variable factors such as wind speed distribution, load growth, electricity tariff, restrictions on operation scheme, and reliability requirements, etc. With each auxiliary solution there is a separate computational model. A preliminary study on an investment of a lowload diesel generator has been conducted and the results are presented here.

The existing diesel generators and the wind turbines belong to two different owners. The investment analysis is carried out for the wind turbine owner, on the basis of maximising profit from increased penetration. The objective function of the model is as follows:

Maximisation (within the project lifetime): Revenue from selling electricity - fuel costs for low-load diesel generator - investment cost for low-load diesel generator.

Principal technical constraints consist of:

- Balancing active and reactive power.

- Constraints on generation characteristics of lowload and existing diesel generators (maximum and minimum power).

- Spinning reserve level.

- Maximum allowed penetration for wind power (maximum of 85% in the presence of a low-load diesel generator).

The optimisation model was calculated with different electricity tariffs and with assumptions of different load growth scenarios. Typical results are shown in Figs.6 and 7.

Fig.6 shows the optimal capacity for the low-load diesel generator for each electricity tariff and load growth with the project lifetime of 5 years. The diesel generator is responsible for 100% spinning reserve. The results clearly show that it is efficient to invest in a low-load diesel generator of 300kW. Once the tar-



Fig.6. Investment analysis for a low-load diesel generator. Diesel generator responsible for 100% spinning reserve

iff reaches 14US cents/kWh, the investment proves cost-effective.

In case the diesel generator is responsible for 90% spinning reserve and all other assumptions are kept unchanged, the results are shown in Fig.7. In comparison with Fig.6, the financial benefits in this case become evident. Both analyses show that when the requirement for reliability can be compromised (higher risks), the investment for a low-load diesel generator yields a better return.

5. Conclusions

This paper reports technical and financial challenges that need resolving for the existing wind-diesel hybrid system in Phu Quy island.

In order to improve the system stability and reliability the following technical solutions are recommended:

- Operate 3 diesel generators at low and medium loads, 4 diesel generators at peak load in order to stabilise system frequency.

- Allow wind penetration level up to 70% on the basis of diesel generators covering all of the spinning reserve.

- Consider the possibility of using the frequency control function of V80 wind turbines in combination with the control function of the SCADA system to improve frequency stabilisation.

Regarding financial models to invest auxiliary equipment, it is concluded that:

- At present, a low-load diesel generator is considered to be a potential and feasible investment as this solution meets all technical requirements i.e. stability, spinning reserve, increase of wind penetration level and simple operation scheme.

- The solution of using a dump-load and a compensation capacitor is also attractive due to low cost invest-



Fig.7. Investment analysis for a low-load diesel generator. Diesel generator responsible for 90% spinning reserve

ment and it may increase the wind penetration level in certain cases.

- Energy storage devices are considered not necessary at the moment due to low load demand as compared to the total installed capacity. Auxiliary storage equipment can be designed to improve stability and voltage controllability, however costs are high.

- In all cases the solution for the financial model to select appropriate auxiliary equipment depends on a number of factors such as electricity tariff, technical constraints and reliability requirements.

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ENVIRONMENTAL IMPACTS OF BIO-FUELS THROUGH EMISSION

Le Quoc Phong Petrovietnam University

Summary

Air pollution is emerging to become a global problem. Emission from vehicles is one of the most dangerous polluters. Use of bio-diesel is one of the means to reduce the emissions, particularly of particulate matter (PM) and nitrogen oxides (NO_x). The trend toward using bio-fuel as an alternative fuel will significantly increase in the near future as crude oil resources become depleted. This paper presents experimental results from a heavy-duty diesel engine using different bio-diesel fuels. Experimental bio-diesel fuels are rapeseed methyl ester (RME), blends of RME with conventional diesel fuel, gas to liquids (GTL), blends of GTL with conventional diesel fuel, O_2 -diesel fuels called O_2 -1 (O_2 -diesel) in this experiment (a blend of ethanol and diesel fuel) and the blend of O_2 -diesel and 5% RME called O_2 -2 (95% O_2 -diesel - 5% RME). The results presented here demonstrate that those bio-fuels not only represent a solution to counter the mounting scarcity of fossil fuels but also reduce the central elements of emissions.

1. Introduction

A substance in the air that can be harmful to humans and the environment is known as an air pollutant. Pollutants can be in the form of solid particles, liquid droplets, or gases. In addition, they may be natural or man-made. Exhaust gases from engines primarily contribute to forming air pollutants [1]. Therefore, finding methods which reduce emissions from vehicles to meet more stringent requirements of limiting air pollution are important and urgent tasks for researchers, manufacturers and regulators.

Besides technological solutions to reduce emissions from internal combustion engine, bio-fuels and their blend with fossil fuel have been used for a long time as they seem to be the most promising solution to emissions and the depletion of crude oil resources. Bio-fuel counterparts can be bio-gasoline (bio-ethanol), bio-diesel and bio-jet fuel. In practice, ethanol or the blend of ethanol and conventional fuels have usually been used for gasoline engine due to their suitable properties [1]. Therefore, researching the effects of the mixture of diesel fuel and ethanol as oxidised substance is an experimental goal. In this paper, the blend of ethanol and conventional diesel fuel called O₂-diesel (manufactured by Shell) or O₂-1 and the blend of O₂-diesel and 5% RME called O₂-2 are used to experiment because of the increasing trend of using diesel vehicles in transportation. In our experiments, the use of other bio-diesel such as RME (B100), the blend of RME and diesel fuel (10% RME - B10, 50% RME - B50), GTL (G100) and the blend of GTL and diesel fuel (50% GTL- G50) are also studied. Experiments have also been carried out at different operating parameters such as full load, part load of engine and different EGR ratios (Exhaust Gas Recirculation - Exhaust gas is recirculated into the intake manifold of engine to reduce maximum flame temperature in the combustion chamber) to generally and deeply analyse the emission of bio-diesel.

2. Emission and environmental and social health effects

The combustion of fuels with air containing O_{γ} , inert gases and N, releases energy as heat in an exothermic reaction [1]. The heat released in internal combustion engine by hydrocarbon-based fuels such as gasoline and diesel fuel is determined by numerous incomplete reactions dependent on the compositions of the hydrocarbons in the fuel. Diesel fuel is composed of about 75% saturated hydrocarbons (primarily paraffins including n, iso, and cycloparaffins), and 25% aromatic hydrocarbons (including naphthalenes and alkylbenzenes) [1, 2]. Besides the main components such as CO₂, water vapourvapour and N₂, pollutants such as carbon monoxide, unburned and partly burned hydrocarbons (HC-aldehydes, ketones, etc.) and NO, also exist in exhaust gas of a diesel engine. These pollutants must follow national and international regulations. Pollutants are primarily produced by an interruption of the reaction chain when its dwell time in the combustion chamber is short. Hence, the equilibrium no longer exists. Inhomogeneities in the mixture caused by different air-fuel ratios (λ), combustion chamber wall effects and impurities and additives in the fuel also contribute to undesired by-products. Depending on the type of the fuel and combustion process, solids may also be present as particle emissions. Unrestricted exhaust components produced by the thermal cracking of hydrocarbons and its by-products are increasingly attracting attention since they either are potentially hazardous or cause a discernible odour [3].

2.1. Nitrogen oxides (NO_x)

Unlike CO and hydrocarbon, $NO_{x'}$ which consists of nitrogen monoxide (NO) and nitrogen dioxide (NO_2), is a product of complete combustion. Nitrogen monoxide is the predominant nitrogen oxide produced inside an engine cylinder. The principal source of NO is the oxidation of atmospheric (molecular) nitrogen. However, if the fuel contains a significant amount of nitrogen, the oxidation of the fuel compounds containing nitrogen is an additional source of NO. It is generally accepted that the following are the principal reactions in the combustion of near stoichiometric fuel-air mixtures. Often called the extended Zeldovich mechanism, these reactions govern the formation (and destruction) of NO from molecular nitrogen [1]:

$$O + N_2 \rightarrow NO + N$$

 $N + O_2 \rightarrow NO + O$
 $N + OH \rightarrow NO + H$

NO forms in both the flame front and post-flame gases. However, since combustion in engines occurs at high pressure, the flame reaction zone is extremely narrow (\approx 0.1mm) and the residence time within the zone is brief. Also, due to the cylinder pressures that increase during the combustion process the gases are compressed to a temperature higher than immediately after combustion. Thus, NO formation in the post-flame gases almost always dominates any formation of NO in the flame-front. Furthermore, the NO formation rate is a function of the gas temperature and fuel-air equivalence ratio in post-flame gases. In an engine, a noteworthy NO level can first be detected above around 1,900°C [1]. Overall, more than 90% of NO₂ emissions are nitrogen monoxide. Fuel nitrogen is also a source of NO by means of a different and not yet fully explained mechanism. The nitrogen in distillate fuels can exist as amines and ring compounds (e.g. pyridines, quinolines and carbazoles). During combustion, these compounds are likely to undergo some thermal decomposition prior to entering the combustion zone. Therefore, the precursors of NO formation are nitrogen-containing compounds with low molecular weights such as $\rm NH_{3'}$ HCN and CN. Detailed information on the kinetics of NO formation from these compounds is limited. A characteristic of NO is its reactivity with oxygen and particularly quick reactivity with ozone to form $\rm NO_2$. Pure NO does not irritate the lungs but if $\rm NO_2$ remains unconverted, methemoglobin forms after resorbed by the respiratory tract. NO is an endogenous modulator of blood vessel tone and thus a well studied substance in terms of physiology and metabolism [1].

 NO_2 has a pungent odour and a reddish brown color. A plausible mechanism for the persistence of NO_2 is:

$$NO + HO_2 \rightarrow NO_2 + OH$$

Chemical equilibrium considerations indicate that NO_2/NO ratios should be negligibly small and NO_2 can be 10% of total exhaust of nitrogen oxides emissions [1, 2]. As a free radical, NO_2 is basically in a position to abstract hydrogen from fatty acids and thus to cause lipid peroxidation, which ultimately leads to a loss of function in biological membranes. In the presence of water, NO_2 develops as a cellular poison in the respiratory tract, through the nitric acid (HNO₃).

2.2. Hydrocarbon (HC)

Ideally, an internal combustion engine's combustion process corresponds to the following reaction [1]:

$$C_n H_m + (n + \frac{m}{4}) O_2 \rightarrow n CO_2 + \frac{m}{2} H_2O$$

In practice, the combustion process does not produce carbon dioxide and water alone. Thus, combustion in an internal combustion engine is usually an incomplete process. As a result, incomplete combustion of the hydrocarbon-based fuel results in hydrocarbons or more organic emissions. The level of unburned hydrocarbon in the exhaust gases is generally specified in term of the total hydrocarbon concentration expressed in parts per million. Engine exhaust gases contain a wide variety of hydrocarbon compounds. Diesel fuel contains hydrocarbon compounds with higher boiling points and hence higher molecular weights than gasoline [1, 2]. Also, substantial pyrolysis of fuel compounds occurs within the fuel sprays during the diesel combustion process. Thus, the composition of the unburned and partly oxidised hydrocarbons in the diesel exhaust is much more complex than in the spark-ignition engine and extends over a larger molecular size range. Hydrocarbon constituents range from methane to the heaviest hydrocarbons that remain in the vapour phase in the heated sampling line (which is usually maintained at about 190°C) [2]. The levels of hydrocarbon emission from diesel engines vary widely depending on operating conditions.

So far it has not been easy to assess the direct impact of hydrocarbon emissions on humans. Hydrocarbon emissions are sometimes slightly relevant toxicologically (alkanes or alkenes etc.) and can also be carcinogenic (benzene) [2]. Hydrocarbons from engine emissions contribute to the development of summer smog.

2.3. Soot

Soot particles form primarily from the carbon in the diesel fuel and the incomplete combustion [1].

$$C_n H_{2n+2} + (n+1)O_2 \rightarrow nC + (n+1)H_2O$$

The formation process begins with a fuel molecule containing 12 - 22 carbon atoms and having an H/C ratio of about 2 and ends with particles that are typically a few hundred nanometres in diameter, composed of spherules with 20 - 30nm in diameters, each of which contains some 10⁵ carbon atoms and has an H/C ratio of about 0.1. Soot forms in the diesel combustion environment at temperatures between around 1,000 and 2,800K, at pressures of 50 - 100 bar and with sufficient air to fully burn all the fuel [2]. Solid soot particles form from a fraction of the fuel in milliseconds. Soot formation occurs in two stages. In the first stage, particles form. The initial condensed phase material is produced by the fuel molecules by means of oxidation and pyrolysis products. These products typically include various unsaturated hydrocarbons, particularly acetylene's higher analogues $(C_{2n}H_2)$ and polycyclic aromatic hydrocarbons (PAH). These two types of molecules are considered the most likely precursors of soot in flames. In the second stage, particles grow. This involves surface growth, coagulation and aggregation. Surface growth, which generates the bulk of the solid-phase material, involves the attachment of gas-phase species to the surface of the particles and their incorporation into the particulate phase [2]. Surface growth reactions lead to an increase in the amount of soot but the number of particles remains unchanged. These stages of particle generation and growth constitute the soot formation process. In each stage, oxidation can occur, during which soot or soot precursors are burned with oxidising species to form gaseous products such as CO and CO₂. The real emission of soot from the engine will depend on the balance between these processes of formation and burnout [1, 2].

2.4. Particulate matter (PM)

Diesel particulates primarily consist of carbonaceous material (soot) generated in combustion, which have absorbed some organic compounds [1]. Most particulate material results from incomplete combustion of fuel hydrocarbons, but also from the lubricating oil. Other sources are hydrocarbon compounds (some of which are bound to soot) and a few sulfates in the form of aerosols. In a combustion engine, the development of particles is related to the development of soot, which in turn basically depends on the local temperature of the zone the fuel enters and a low oxygen supply. According to the Acetylene Theory, the combustion of different hydrocarbons runs through several intermediate substages such as cracking and dehydration. This increases the share of carbon molecules until the first particle with a diameter larger than 0.01µm develops, around which so-called primary particles coagulate to form larger units with diameters from 0.01 - 0.08µm (forming secondary particles). Their large specific surfaces enable uncombusted and partly combusted hydrocarbons, especially aldehydes, to bond to the secondary particles. When combustion occurs, the secondary formation phase is soot reoxidation governed by the dwell time and oxygen concentration. A specific problem of diesel engines is the conflict between particles and NO_v. The conditions for low particle formation and low hydrocarbon emissions (achieved with higher temperatures) conflict with the preconditions for low nitrogen oxide emissions. Therefore, attention is concentrated on the secondary formation phase of soot reoxidation. Soot reoxidation necessitates a large amount of mixture formation energy in the last phase of combustion. This can be attained by a specific swirl and tumble in the combustion chamber, higher injection pressure and a faster injection rate at the end of the injection process. Unfortunately, these conditions are prerequisites to high NO_v emissions.

Numerous environmental studies have concluded that increases of particle concentration in the air correlate with increases in the number of patients with respiratory and circulatory illnesses. Also found is a link between the risk of lung cancer and particles as well as ozone and sulfur dioxide. In general, particle emissions have a negative impact on human health, especially in particularly sensitive individuals, e.g. children, the elderly and invalids.

2.5. Carbon monoxide (CO)

Carbon monoxide is likewise a product of an incom-

plete combustion process. CO emissions from internal combustion engines are primarily controlled by the air fuel ratio. CO concentrations in the exhaust of fuel-rich mixtures steadily increase as the air fuel ratio decreases, i.e. as the amount of excess fuel increases. Since the fuelair mixture must be enriched when an engine is cold, CO emissions during engine warm-up are much higher than emissions in the fully warmed-up state.

Carbon monoxide is a colourless and odourless gas and attaches itself to hemoglobin 250 times more strongly than oxygen. As CO concentrations increase in inhaled air, the suppression of oxygen induces symptoms from suffocation to death. Acute poisoning occurs above 2,000ppm. All in all, CO emissions from internal combustion diesel engines are insignificant [1, 3].

2.6. Unregulated emission components

Important unregulated exhaust gas components in untreated exhaust from diesel engines are cyanide, ammonia (NH_3), sulfur dioxide (SO_2) and sulfates. Of the specific hydrocarbons, methane, ethane, ethene, ethine, benzene and toluene are of particular interest. Of the PAH, phenanthrene, pyrene, fluorene, fluoranthene and anthracene predominate and in that order. The concentration of these components is at least six times higher than that of other individual PAH substances and forms about 90% of the PAH [3]. The components cited are formed from trace substances in the fuel, in the lubricant and, to a degree, from secondary reactions in the exhaust system.

Combustion exhaust gases contain a multiplicity of different toxic elements. The bulk of the over 150 PAH substances identified are mutagenic and carcinogenic. Many studies have been carried out on the influences of diesel engine emissions on the health of humans and the environment [2, 3]. In 1978, an Ames Test (Salmonella Microsome Test) performed by Huisingh demonstrated for the first time the capability of diesel engine emissions to cause genetic damage. Since then, many further studies have confirmed these findings [3].

In this paper, main emissions from diesel engine such as hydrocarbon, NO_x , PM (illustrated through soot or FSN- Filter Smoke Number) with different biofuels and the blends have been evaluated. FSN have been measured at full load with different speeds (from 600rpm-idle to 2,400rpm-maximum speed). In addition, FSN and other emissions such as hydrocarbon and NO_x have been tested at part loads (1,100rpm - 213Nm and 425Nm), full load (1,400rpm - 850Nm) with different EGR (Exhaust Gas Recirculation- the effective method to reduce NO_x emission in diesel engine) ratios (0%, 9% and 27%).

| Parameters | Testing procedure | Unit | DIN EN 590 | Diesel fuel (DF) | O ₂ -diesel |
|---|--------------------|-------------------|-----------------------------|-------------------|------------------------|
| Heat value | DIN 51 900-2 | MJ/kg | approx. 42 | 43.866 | 42.360 |
| Cetane number | DIN EN ISO 5165 | | min. 51 | 55.5 | 54.8 |
| Cetane index | DIN EN ISO 5165 | | min. 46 | 54.6 | 52.8 |
| Density in 15°C | EN ISO 12185 | g/cm ³ | 0.820 - 0.845 | 0.8276 | 0.8252 |
| РАК | DIN EN 12916 | % (m/m) | max. 11 | 2.0 | 1.9 |
| Sulfur content | EN DIN 51400-10 | mg/kg | max. 10 | 8 | 3 |
| Flash point | DIN EN 22719 | °C | > 55 | 56.9 | 12.9 |
| Coke residue (of 10% distillation residue) | DIN EN ISO 10370 | % (m/m) | max. 0.30 | 0.004 | 0.08 |
| Water content | EN ISO 12937 | mg/kg | max. 200 | 130 | 250 |
| Oxide ash | DIN EN ISO 6245 | % (m/m) | max. 0.01 | 0.001 | 0.004 |
| Total contamination | DIN EN 12662 | mg/kg | max. 24 | 27.3 | 27.8 |
| Copper corrosion (3 hours at 50°C) | EN ISO 2160 | | 1 | 1 | 1 |
| HFFR Lubricity with 60°C | DIN EN ISO 12165-1 | μm | max. 460 | 317 | 284 |
| Viscosity in 40°C | DIN 53015 | mm²/s | 2.00 - 4.50 | 2.71 | 2.03 |
| Distillation % [V/V] - 250°C % [V/V] - 250°C 95% - Point | DIN EN ISO 3405 | Vol% Vol% ℃ | < 65 min. 85 max. 360 | 41 94 352.9 | 45 95 353.3 |
| Fatty acid methyl ester content | EN 14078 | % (V/V) | max. 5 | 0.17 | 0.16 |
| Cold filter plugging point (CFPP) | DIN EN 116 | °C | max.0 | -8 | -7 |

Table 1. Properties of O₂-diesel (ethanol-diesel blend) [5]
3. Experimental bio-diesel

3.1. O_2 -diesel (Ethanol diesel fuel) or O_2 -1(as called in this experiment)

 O_2 -diesel that is used in this study is supplied by Shell Company. The analysis of O_2 -diesel used for the experiments in this study was performed at the University of Rostock. Table 1 presents the properties of experimental O_2 -diesel.

O₂-diesel fuel is a mixture that consists of the following components [5]:

Water-free ethanol: 7.7% Vol.

Diesel fuel: 91.6% Vol.

Additive 2EHN: 0.2% Vol.

Additive O2DO5: 0.5% Vol.

The additive 2EHN with 0.2% Vol. was used to enhance the cetane number for a number of reasons. One of them is that a small amount of water can be introduced into the fuel system because ethanol has an affinity to water (the experimental ethanol diesel blend had a water content of less than 0.5%) and because blend levels of up to 15% are often desired. One way to achieve this is to use additives. Two common types of additives are surfactants and co-solvents. O₂-diesel with these additives are very stable. Co-solvents have an immediate polarity between ethanol and diesel fuel and act as a bridging agent to produce a homogeneous blend.

Experiments were carried out with O_2 -1 and the blend of O_2 -diesel and 5% RME (called as O_2 -2 in this experiment).

3.2. Rapeseed methyl ester (RME)

RME is manufactured from rapeseed. RME has been used as bio-diesel fuel in Europe for a long time. Since the 2,000s, diesel fuel, including 5% bio-diesel (RME), is used in practice [4]. Therefore, experiments are being conducted with different blending ratios of diesel fuel and RME, for example 100% RME, B50 (50% RME), B10 (10% RME). Table 2 show the properties of experimental RME.

Table 2. Properties of RME [5]

| Parameters | Testing procedure | Unit | Diesel fuel | RME |
|--|----------------------|-------------------|----------------|---------------|
| Heat value | DIN 51606 | MJ/kg | 43.866 | 37.6 |
| Cetane number | DIN EN ISO 5165 | | 55.5 | 45 - 59 |
| Density at 15°C | ISO 3675 | g/cm ³ | 0.8276 | 0.875 - 0.900 |
| Sulfur content | ISO 4260 | mg/kg | 8 | 9 - 12 |
| Flash point | ISO 2719 | °C | 56.9 | 110 |
| Coke residue (of 10% distillation residue) | DIN EN ISO 10370 | % | 0.004 | 0.05 |
| Water content | DIN 51606 | mg/kg | 130 | 300 |
| Oxide ash | DIN EN ISO 6245 | %mass | 0.001 | 0.01 |
| Total contamination | DIN EN 51419 | mg/kg | 27.3 | 20 |
| Copper corrosion (3 hours at 50°C) | EN ISO 2160 | | 1 | 1 |
| Viscosity at 40°C | ISO 3104 | mm²/s | 2.71 | 3.5 - 5 |
| Cold filter plugging point (CFPP) | DIN EN 116 | °C | -8 | -9 to -7 |

Table 3. Properties of GTL [5]

| Parameters | Testing procedure | Unit | Results |
|-----------------------------------|-------------------|-------------------|---------|
| Doncity | EN ICO 12195 | a/cm ³ | 0 7052 |
| Defisity | | g/cm | 0.7635 |
| Viscosity at 40°C | DIN EN ISO 3104 | mm²/s | 3.58/1 |
| Cloud point | DIN EN 23015 | °C | 0 |
| Cold filter plugging point (CFPP) | DIN EN 116 | °C | -3 |
| Sulfur content | DIN 51400 T11 | mg/kg | < 5 |
| Heat value | ASTM D240 | MJ/kg | 44,398 |
| Cetane number | | | 74 |
| Distillation | DIN EN ISO 3405 | | |
| Boiling point | | °C | 202.3 |
| T10 | | °C | 244.7 |
| T50 | | °C | 295.2 |
| Т90 | | °C | 340.9 |
| Boiling end point | | °C | 351.3 |
| Residue | | % v/v | 2.0 |
| Flash point | ASTM D93 | °C | 89 |
| Carbon to hydrogen ratio | | | 2.13 |
| Aromatics | ASTM D5186 | mass % | 1.4 |
| Gum content | ASTM D381 | mg/100ml | 5.9 |
| Ash | ASTM D482 | mass % | < 0.001 |
| Carbon residue | ASTM D524 | mass % | 0.03 |

3.3. Gas to liquids (GTL)

Produced by the Shell Middle Distillate Synthesis (SMDS), GTL is one of the new alternative fuels used for diesel engines without any modifications. The Lab of Shell Global Solutions (Deutschland) GmbH in Hamburg performed the testing of the GTL properties. Table 3 presents the test results.

Experiments are conducted with different blending ratios of diesel fuel and GTL, for example 100% GTL, G50 (50% GTL).

4. Effects of bio-diesel and these blends on emission

Research was carried out on a DEUTZ BF6M 1013 EC

heavy-duty engine with different operating points. The DEUTZ BF6M 1013 EC is a four-stroke direct injection engine with six in-line cylinders and turbocharger and are retrofitted with exhaust gas regulations (EURO II). Experiments have been carried out in the laboratory of the Institute of Mobile Systems, University of Magdeburg, Germany. Experiments have also been conducted at the different operating points of engine such as full load, part load and different EGR ratios.

At full load of engine, diesel fuel, O_21 , B10 (10% RME - 90% DF), B50 (50% RME - 50% DF), B100 (100% RME) and GTL (G100) were tested. At other operating points of the engine, diesel fuel, O_2 -1, O_2 -2, B10, B50, B100 (RME) were G50 (50% GTL-50% DF), G100 were experimentally examined.

Besides the different operating points, experiments were also carried out at different EGR (Exhaust Gas Recirculation) ratios. EGR is the effective method to reduce NO_x emission in diesel engine. EGR has not been analysed in detail in this paper.

4.1. FSN

Bio-diesel and their blends point to better emissions trends, especially the FSN at full load (Fig.1). The FSN decreases by roughly 40% with B100 and at least 21% with O_2 -diesel and B10. The FSN falls around 38% with B50. GTL 100% has a lower FSN than O_2 -diesel, except at speeds above 1,800rpm. Compared with diesel fuel, GTL 100% decreases by about 6.4% in the minimum point (2,200rpm) and around 22% in the maximum point (800rpm). All the alternative fuels and their blends have generally been found to decrease the FSN.

Experimental results also confirmed that fuel temperatures had less impact on engine torque and fuel consumption (B g/h) when RME, G100 and diesel fuel were used than when O₂-diesel was used. The temperatures of RME, GTL and conventional diesel fuel were similar under most operating points and conditions. By contrast, the temperature of O₂-diesel significantly influences engine torque and fuel consumption. Experimental results indicate the engine runs unstably when the temperature of O₂-diesel is around 55°C (the same working temperature as diesel fuel, GTL and RME). When the temperature increased from around 30 - 55°C, engine torgue changed approximately 10% and B_o approximately 8.3%. The evaporation of ethanol in O₂-diesel and its lower boiling temperature range are responsible for these effects. Experimental results also confirmed the engine ran stably

when the temperature of O_2 -diesel was approximately 30°C. Therefore, this was the temperature maintained in experiments with O_2 -diesel.

Fig.2 presents decreasing trends of FSN for all alternative fuels and different operating modes of engine, compared with diesel fuel, except when the EGR is 27% and except for G100 and G50. The changing percentages are largest with B100 and lowest with GTL, its blend and B10. The maximum changing percentage is over 60% with B100 at lower part load (1,100 1/min - 213Nm). When the speed is the same (1,100 1/min), EGR is 9% and engine load increases from 213 - 425Nm, the changing percentage decreases from over 50% to over 40% with B100 and increases modestly with O₂-diesel.

4.2. NO_x

Compared with diesel fuel, the largest increased percentages are always found with B100 and the strongest decreased percentages usually occur when G100 is used (Fig.3) because RME has the lowest heat value, the highest density, highest injection mass and highest injection pressure not only compared with diesel fuel but also other alternative fuels. Therefore, the combustion rate and combustion temperature peak are highest with RME.







Fig.2. Change of FSN in percentage, compared with diesel fuel [5]



Fig.3. Change of NOx in percentage, compared with diesel fuel



Fig.4. Change of hydrocarbon in percentage, compared with diesel fuel

Compared with diesel fuel, the largest increased percentage is over 30% with B100 when EGR is 27%. The strongest decreased percentage is over 20% with G100 under the same conditions (Fig.3). RME blends have lower changing percentages than neat RME and G50 has lower changing percentages than G100. Changing percentages of NO_x emission of different fuels are smaller when engine load increases. When the speed is maintained (1,100 1/min), there is no EGR and the engine load increases from 213 -425Nm, the changing percentage of NO_x with B100 drops from 24 - 18% and is around 6.7% when the engine runs at maximum torque (1,400 1/min - 850Nm).

Generally, GTL and the blend always present a decreasing trend of NO_x emission. In contrast, other alternative fuels and their blend such as RME and O_2 -diesel show an increase of NO_x emission. Therefore, a combination of EGR and bio-fuel is used for reducing NO_x emission in diesel engines.

4.3. Hydrocarbon

Hydrocarbon emission changes significantly with B100, G100 and G50 (Fig.4). Maximum changing percentages (over 50%) occur with B100 and G100 when the EGR ratio is about 9% and the engine runs at higher part load. Compared with diesel fuel, the maximum increased percentages are over 20% (about 8ppm) with B10 at maximum torque and without EGR and over 30% (over 30ppm) with O_2 -diesel at higher part load and without EGR.

Like NO_x emission, hydrocarbon also increases when engine runs with GTL and O_2 -diesel. Higher increase occurs with higher load of engine.

5. Conclusions

Emissions decrease with most bio-fuels. Changing percentage of emission increases with higher content of biofuel that is mixed with conventional fuel. Combination of other methods such as EGR, load, EGR temperatures, and bio-fuels are positive means for decreasing pollutants from the exhaust gas of an diesel engine. This confirmed that use of bio-fuels or the blend of bio-fuels and conventional fuels will meet stricter future requirements for emissions which pollute the environment. The experimental results also confirmed that step-by-step increase of the percentage of bio-fuel in the blend is a suitable practical method in applying progress. Finding and applying bio-fuels will be important future goals for the world. Protecting and encouraging bio-fuel use through tax and other policies will be required for a green planet in the future.

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PETROVIETNAM NEWS

Vietnam, Kuwait beef up energy co-operation

On 22 October 2013 in Ha Noi, Prime Minister Nguyen Tan Dung received Kuwaiti Deputy Prime Minister and Minister of Oil Mustafa Al-Shemali. Prime Minister Nguyen Tan Dung said Vietnam always regards Kuwait as one of its key economic partners and hopes to further foster linkages, particularly in the field of oil and gas. The Prime Minister highly appreciated the Kuwaiti Government's role in speeding up the implementation of the Nghi Son Refinery and Petrochemical Complex Project, especially its commitment to supply crude oil for the complex.

Kuwaiti Deputy Prime Minister Mustafa Al-Shemali said he was pleased to witness the ground breaking of the complex, which is a joint venture project between Kuwait, Japan and Vietnam. Vietnam and Kuwait, said the guest leader, own vast potentials for further co-operation. Kuwait wants to boost co-operation with Vietnam in the energy, industry and petroleum



Prime Minister Nguyen Tan Dung and Kuwaiti Deputy Prime Minister Mustafa Al- Shemali. Photo: Nhat Bac

sectors, said Mr. Mustafa Al-Shemali while promising to create favourable conditions for Vietnamese investors to do business in Kuwait and stand ready to support human resource training in the oil and gas field.

Nguyen Hoang

The 10th ASCOPE Conference & Exhibition to be hosted by Petrovietnam for the first time



The 10th ASCOPE Conference & Exhibition will take place from 28 to 30 November in Ho Chi Minh City. Photo: PVN

The 10th ASCOPE (ASEAN Council on Petroleum) Conference & Exhibition (ASCOPE C&E) entitled "Innovation and Co-operation - The way forward" will take place in Ho Chi Minh City from 28 to 30 November, 2013. Hosted by Petrovietnam for the first time, this event is expected to showcase about 300 exhibitors and bring together approximately 500 conference delegates. Leading companies in the oil and gas industry will also be there to exhibit up-to-date tools and technologies for oil and gas exploration, production and transportation, equipment for energy resources metering and control, automatic process control systems, integrated operational systems for plant management, software, innovative projects for oil and gas facilities construction, a wide variety of fittings, protective equipment, and services for oil and gas complexes, etc.

The conference in particular brings a unique opportunity to effectively discuss burning issues of the oil and gas industry. The Presidents and CEOs of all ASEAN national oil companies are expected to make keynote speeches during the conference and they will be joined by a host of other senior executives from the region's oil industry. Together, they will provide a timely insight into the ASEAN petroleum business, challenges to be faced, strengths to be enhanced and solutions to be applied. In addition, there will be a technical conference for the region's petroleum engineers.

Nam Rong - Doi Moi oil field reaches oil production milestone of 1 million tons



Nam Rong - Doi Moi oil field. Photo: PVEP

On 27 September 2013, the project partners in the Nam Rong – Doi Moi oil field production project celebrated its production of 1 million tons of crude oil. Attending the ceremony were leaders of Ba Ria - Vung Tau province, the Vietnam Oil and Gas Group (Petrovietnam), Vietsovpetro, VRJ, Zarubezneft, PVEP and Idemitsu.

The structure of Nam Rong - Doi Moi lies in the area of two blocks with separate oil and gas operation permits in the Cuu Long basin on the continental shelf of Vietnam. In 2009, Petrovietnam and its partners signed an Integration and Joint Development contract, merging the Nam Rong and Doi Moi fields into one named Nam Rong-Doi Moi field and appointing Vietsovpetro as the operator. In just 15 months of development, with Vietsovpetro's strong capacity as an operator, the support of ministries and agencies concerned together with Petrovietnam's close guidance and the co-operation of the participating parties including PVEP, the Nam Rong - Doi Moi Project produced its first oil in late 2009. It was put into commercial production in early 2010, saving nearly 2 million USD over the approved budget for field development. After 3 years, 8 months and 4 days, with production maintained at an optimum level and further investments made to complete in-field pipes, the field has reached the oil production milestone of 1 million tons.

In a speech delivered at the ceremony, Chairman of Petrovietnam Dr.Sc. Phung Dinh Thuc noted the efforts of the collective staff, who are working day and night at the field. He further expressed his hopes that with the achieved results, the partners would keep up their efforts to meet the production plan, thus contributing to ensuring that the oil production output of Petrovietnam would exceed 16 million tons this year.

Dinh Nhan

Load-out of Su Tu Vang North East Topside

On 6 October 2013, the Petroleum Equipment Assembly & Metal Structure Joint Stock Company (PVC-MS) successfully loaded out the Su Tu Vang North East Topside at Vietsovpetro port. This is a project invested by Cuu Long JOC, with Vietsovpetro acting as the main contractor and PVC-MS as the fabrication contractor.

Su Tu Vang North East Topside weights over 570 tons. This is part of the Su Tu Vang North East Production Platform fabrication project to be executed by PVC-MS. After more than 4 months of construction and fabrication, PVC - MS has fully controlled the work safety as well as ensured the work progress and quality. The work has been handed over to Vietsovpetro to prepare for offshore installation.

Earlier, on 30 September, PVC-MS had successfully launched BK 17 Topside after nearly 4 months of fabrication. This project is a lightweight rig at Bach Ho field. PVC-MS performed the fabrication and installation of the



Su Tu Vang North East Topside successfully loaded out. Photo: PVC

topside and the technology systems, and connected the upper block with the bearing beams. The total weights of this topside and pipe technology systems are about 360 tons. Vietsovpetro has checked and accepted the project which met the quality and safety requirements and was ahead of its planned schedule.

Minh Phuong

PVFCCo excellently completes the periodic overall maintenance of Phu My Fertiliser Plant

Petrovietnam Fertiliser and Chemicals Corporation (PVFCCo) announces that the periodic overall maintenance of Phu My Fertiliser Plant has been successfully completed ahead of schedule. The overall maintenance of Phu My Fertiliser Plant is carried out once every 2 years. This year, it started from 5 September 2013 and satisfied all four criteria of safety, quality, progress and cost. The plant officially released the urea product with 100% capacity at 6:13 hours on 29 September 2013, which meant 38 hours ahead of schedule. This progress brought benefit worth tens of billions of dongs to PVFCCo and contributed to the timely supply of fertilisers for the next winter-spring crop.

Reportedly, in the 4th quarter of 2013, demand for fertilisers throughout the country is expected to be over 500 thousand tons while supply of fertilisers from domestic enterprises is estimated to reach 570 thousand tons, in which Phu My Fertiliser Plant's output is approximately over 200 thousand tons. Phu My Fertiliser Plant, which



Phu My Fertiliser Plant. Photo: DPM

is owned by PVFCCo, has the capacity of 800,000 tons a year. It is using leading advanced technology provided by Haldor Topsoe to produce ammonia and urea production technology provided by Snamprogetti (Italia).

Thuy Nguyen

Inauguration ceremony held for Hua Na Hydropower Plant



Hua Na Hydropower Plant will produce 717 million kWh per year. Photo: CTV

On 20 September 2013, the Petrovietnam Power Corporation (PV Power) organised the inauguration ceremony for the Hua Na Hydropower Plant in Que Phong District, Nghe An province. Participating and witnessing this event were Minister of Industry and Trade Vu Huy Hoang, Chairman of People's Committee of Nghe An province Nguyen Xuan Duong, and the leaders of Petrovietnam. With a total investment of 7 trillion VND (329 million USD), the Hua Na Hydropower Plant with a designed capacity of 180MW and two generators has been completed on schedule, bringing huge economic benefits. The plant will supply an average of 717 million kWh per year to the national electricity grid. It will also contribute to controlling floods in the lowlands and supply water for the winter-spring crop of the western rural districts of Thanh Hoa province.

The Hua Na Hydropower Plant accounts for nearly 40 percent of the total capacity of Nghe An province's power system, thus having an important significance in the socio-economic de-

velopment of the province and contributing to ensuring the national energy security. This plant at the same time marked a significant step in the progress of investors and contractors to complete a major project of Petrovietnam on schedule, especially with all steps from management, design and construction to operation being undertaken by Vietnamese people. Thuy Hang

PV Drilling and UMW supply service jack-up rig to JVPC



Representatives of PV Drilling, UMV and JVPC signed the contracts. Photo: PVD

On 9 October 2013, a signing ceremony was held in Ho Chi Minh City for the main contract between Petrovietnam Drilling and Well Services Corporation (PV Drilling) and JVPC, and a subcontract between PV Drilling and UMW for the provision of jack-up rig NAGA 3 to JVPC's 2014 drilling campaigns. Besides, PV Drilling will provide services for JVPC in Block 15-2 offshore Vietnam.

Along with efficient operation of its wholly owned drilling rigs, PV Drilling is co-operating with international oil contractors to provide services to oil companies that are operating in Vietnam. This is the new opportunities for PV Drilling to enhance its professionalism in drilling rig services provision and increase its market share for this type of service in Vietnam.

With a vision of becoming an internationally reputable and reliable drilling contractor and drilling-related service provider in the oil

and gas industry, PV Drilling has been striving to show continuous improvement in its service quality and increasing its efficiency while maintaining the highest levels of safety and value for its clients.

Nhon Trach 2 Power Plant reaches commercial electricity output of 10 billion kWh

At 18:55 hours on 25 September 2013, the Nhon Trach 2 Power Plant reached power output of 10 billion kWh after nearly 2 years of commercial operation only. This is a proud achievement for a power plant of such capacity and also a milestone marking the success of the Nhon Trach 2 Power Plant in contributing to ensuring the national energy security.

Following the achievements made in recent years, Petrovietnam Power Nhon Trach 2 Joint Stock Company (PVPower NT2) has strived to ensure the safe and efficient operation of the Nhon Trach 2 Power Plant in the competitive electricity market, with all its

planned targets being exceeded, positively contributing to ensuring energy security. The Nhon Trach 2 Power Plant, with a capacity of 750MW and the ability to supply 4.2 billion kWh per year to the national grid and to col-



Nhon Trach 2 Power Plant. Photo: CTV

lect a revenue equivalent to 5,000 billion VND per year, is an important project in the national power system, especially in the Southeast region of Vietnam.

Nguyen Hang

PETROVIETNAM



OIL & GAS GLOBAL MARKET

Worldwide oil demand and supply

the According to the projection by International Energy Agency, global oil demand this year will average 90.6 million b/d. Consumption declines in many of the OECD countries have been larger than expected. OECD demand averaged 45.2 million b/d in the first half, down from 45.9 million b/d last year. Total oil consumption in non-OECD countries is forecast to average 45.1 million b/d for the year. Among these economies, China will post the largest demand growth and its oil demand will climb to average 10 million b/d in 2013, up 4% from 2012. Average demand in the former Soviet Union is to rise by 100,000b/d this year to 4.6 million b/d.

Worldwide oil supply will average 91.8 million b/d this year, led by an increase in OECD America and non-OECD output. IEA expects total OECD crude oil supply to be up 88,000b/d in 2013 at 20.7 million b/d, among this North American OECD Table 1. World oil balance evolution from 2011 to 2013 (million b/d)

| | 2013 2012 | | | | | | 2011 |
|----|--------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| No | Demand/Supply | 1 st | 4 th | 3 rd | 2 nd | 1 st | 3 rd |
| | | Quarter | Quarter | Quarter | Quarter | Quarter | Quarter |
| I | Total demand | 89.26 | 89.84 | 89.05 | 88.62 | 88.73 | 89.21 |
| 1 | Total OECD | 45.60 | 46.12 | 45.84 | 45.44 | 46.21 | 46.62 |
| | U.S. & Territories | 18.78 | 18.70 | 18.89 | 18.85 | 18.69 | 19.14 |
| | Canada | 2.29 | 2.38 | 2.35 | 2.23 | 2.19 | 2.26 |
| | Mexico | 2.11 | 2.24 | 2.11 | 2.13 | 2.10 | 2.18 |
| | Japan | 5.07 | 4.84 | 4.47 | 4.28 | 5.27 | 4.82 |
| | South Korea | 2.31 | 2.37 | 2.26 | 2.23 | 2.34 | 2.30 |
| | France | 1.75 | 1.71 | 1.73 | 1.71 | 1.81 | 1.75 |
| | Italia | 1.28 | 1.33 | 1.38 | 1.36 | 1.34 | 1.43 |
| | United Kingdom | 1.48 | 1.47 | 1.49 | 1.53 | 1.52 | 1.57 |
| | Germany | 2.30 | 2.44 | 2.41 | 2.39 | 2.32 | 2.41 |
| | Other OECD Europe | 6.33 | 6.69 | 6.78 | 6.77 | 6.68 | 6.95 |
| | Australia & New Zealand | 1.28 | 1.31 | 1.27 | 1.27 | 1.26 | 1.19 |
| | Other countries | 0.62 | 0.64 | 0.7 | 0.69 | 0.69 | 0.62 |
| 2 | Total non-OECD | 43.66 | 43.72 | 43.21 | 43.18 | 42.52 | 42.59 |
| | China | 10.62 | 10.59 | 9.93 | 10.09 | 10.32 | 10.07 |
| | FSU | 4.86 | 4.86 | 4.87 | 4.70 | 4.68 | 4.78 |
| | Non - OECD Europe | 0.70 | 0.72 | 0.72 | 0.70 | 0.69 | 0.74 |
| | Other Asia | 10.61 | 10.49 | 10.23 | 10.68 | 10.42 | 10.40 |
| | Other non-OECD | 16.87 | 17.06 | 17.46 | 17.01 | 16.41 | 16.60 |
| П | Total supply | 88.83 | 89.35 | 89.00 | 89.14 | 89.17 | 88.47 |
| 1 | OECD supply | 23.33 | 23.09 | 22.08 | 22.49 | 22.64 | 22.38 |
| | U.S. | 11.70 | 11.71 | 11.00 | 10.94 | 10.85 | 10.67 |
| | Canada | 4.22 | 4.01 | 3.77 | 3.80 | 3.89 | 3.80 |
| | Mexico | 2.93 | 2.92 | 2.94 | 2.95 | 2.94 | 2.94 |
| | North Sea | 3.00 | 2.90 | 2.76 | 3.20 | 3.37 | 3.34 |
| | Other OECD | 1.48 | 1.55 | 1.61 | 1.60 | 1.59 | 1.63 |
| 2 | Non-OECD supply | | | | | | |
| | FSU | 13.54 | 13.50 | 13.37 | 13.37 | 13.43 | 13.32 |
| | China | 4.46 | 4.51 | 4.41 | 4.36 | 4.37 | 4.26 |
| | Other non-OECD | 11.93 | 12.46 | 12.54 | 12.21 | 12.19 | 12.80 |
| 3 | Total non-OECD and non-OPEC | 29.93 | 30.47 | 30.32 | 29.94 | 29.99 | 30.38 |
| 4 | OPEC | 35.57 | 35.79 | 36.60 | 36.71 | 36.54 | 35.71 |

Source: Oil & Gas Journal, 23 September 2013

countries account for 16.9 million b/d. Non-OECD output will average 29.7 million b/d, compared with 29.5 million b/d in 2012. Total oil supply from non-OPEC countries is forecast to be 54.5 million b/d this year, compared with 53.4 million b/d last year. Total oil supply from OPEC, including 6.6 million b/d of natural liquids, will average 37.3 million b/d, compared with 37.6 million b/d in 2012.

Crude and gas prices

Global liquid fuels supply averaged 90.6 million b/d in July and August 2013, up about 1.2 million b/d from the same two months in 2012, mainly due to rising production from countries outside OPEC, according to a report from the U.S. Energy Information Administration. But the world oil market also turned tighter, due to unexpected supply disruptions from unrest in the Middle East and North Africa occurring at a time of low surplus production capacity.

Commercial inventories in the U.S. and other OECD countries were roughly equal to their year-ago levels, averaging 2.685 billion barrels in July and August. OPEC accounted for most of the outages, with 2.0 million b/d disrupted in the two months mentioned above, mainly

due to deteriorating security and production conditions in Libya, Iraq and Nigeria.

From August 2013, oil and gas prices remain to upside. Strong upward pressure on prices is being supplied by intensifying conflicts in the Middle East, exacerbated supply disruptions from members of the Organisation of Petroleum Exporting Countries (OPEC), upstream field maintenance, and pre-stocking requirements from new refinery capacity. The news about U.S. deliberations over military intervention in Syria in September led NYMEX futures into a choppy session. October crude traded between 106.75USD/bbl and 108.75USD/bbl. ICE October Brent settled 1.15USD lower at 114.01USD/bbl after having dipped to 113.66USD/bbl. Due to the relative releasing tension in Syria at the end of September, the WTI prices were established at 103.31USD/bbl and the Brent at 109.00USD/bbl on 3 October 2013 (Table 2).

On the consumption countries' side, Japan imported 172,047b/d of crude oil from Iran in July, up 33.8% from 128,544b/d in June, according to Japan's Ministry of Economy, Trade and Industry (METI). In July, Japan imported a total 3.49 million b/d, up 1.8% from a year

| Exporters | June | July | August | September | October (forecast) |
|-----------------------------|--------|--------|--------|-----------|-----------------------|
| OPEC reference basket | 101.03 | 104.45 | 107.52 | 106.33 | 106.80 |
| WTI - USA | 99.37 | 104.51 | 106.00 | 108.70 | 103.31 |
| Brent - North Sea | 102.92 | 107.96 | 111.75 | 109.00 | 108.80 |
| Arabia light - Saudi Arabia | 101.30 | 105.03 | 108.09 | 107.28 | 107.09 |
| Basrah light - Iraq | 98.84 | 103.24 | 106.07 | 104.78 | 104.59 |
| Bonny light - Nigeria | 106.12 | 110.21 | 113.62 | 112.39 | 112.18 |
| Es Sider - Lybia | 103.07 | 107.91 | 111.07 | 109.16 | 108.96 |
| Girassol - Angola | 104.23 | 107.55 | 110.80 | 110.39 | 110.18 |
| Iran heavy | 100.61 | 103.65 | 107.06 | 106.55 | 106.36 |
| Kuwait export | 100.22 | 103.22 | 106.47 | 106.14 | 105.94 |
| Marine - Qatar | 102.40 | 103.34 | 106.67 | 106.12 | 105.92 |
| Merey - Venezuela | 95.37 | 95.68 | 98.06 | 100.93 | 100.74 |
| Murban - UEA | 102.61 | 105.58 | 109.18 | 108.67 | 108.47 |
| Oriente - Ecuador | 96.01 | 99.54 | 98.24 | 101.68 | 101.49 |
| Algeria Blend 44° | 102.07 | 107.56 | 111.87 | 108.67 | 108.47 |
| Minas 34º - Indonesia | 103.19 | 103.38 | 105.55 | 109.28 | 109.08 |
| Fateh 32° - Dubai | 100.32 | 103.52 | 106.81 | 106.24 | 106.05 |
| Istmuth 33° - Mexico | 104.08 | 109.18 | 109.09 | 110.23 | 110.02 |
| Urals - Russia | 102.74 | 108.06 | 110.75 | 108.81 | 108.61 |
| WTI/Brent | -3.55 | -3.45 | -4.72 | -0.30 | -5.49 |
| Brent/Dubai | 2.60 | 4.44 | 4.46 | 2.76 | 2.75 |

Table 2. Oil average prices from June 2013 to October 2013 (USD/b)

Source: Oil & Gas Journal, June - October 2013

earlier, marking the second consecutive year-on-year increase in its monthly crude imports, according to METI data. Saudi Arabia remains the top crude supplier to Japan, supplying 1.07 million b/d, which accounted for 30.6% of the country's total crude imports in the month.

About the regional gas market, in Europe, Ukraine plans to reduce imports of Russian natural gas yet further as it broadens its base of energy suppliers, lowering its costs in the process. European gas is 70 - 80USD/1,000m³ cheaper than Russian gas. Ukraine expects to pay 416USD/1,000m³ for Russian gas in the fourth quarter, considerably higher than the price of gas from Europe.

We know that the price of oil is fluctuating and the gas price formula is linked to oil. According to Oil and Gas Journal (2 September 2013), Ukrainian gas importer Naftogaz Ukrainy plans to import just 18 billion m³ of Russian gas in 2013, down from 24.9 billion m³ in 2012. In September 2013, Ukraine signed an agreement that would open imports of European gas via Slovakia. While Ukraine has been importing a small amount of European gas via Hungary and Poland, opening the Slovakian route would be a game changer because of its massive capacity-up to 20 billion m³ of gas each year.

The main suppliers for the Western Europe gas market are African export countries. Sonatrach's full contract volumes are 19.5 billion m³/year to Eni, 6 billion m³/year to Enel and 2 billion m³/year to EDFowned Edison, but it has been a long time since these were delivered in full. Eni bought 14.45 billion m³ from Sonatrach in 2012 only. For Sonatrach, sending less gas to Italy allows it to boost global LNG sales - although it has this year increased piped gas exports to Spain, its other key European market. Uniquely among major LNG exporters, Sonatrach can alter the balance of LNG and piped gas sales, depending on demand. Obviously, the Algerian company can in any case ill-afford to alienate Italian buyers. Eni's contract expires at the end of 2019, but there is little evidence that Sonatrach has made any concessions on oil-indexed pricing of its terms contracts. Because Italy remains a premium market for Sonatrach, it is a wonder if Sonatrach may even have allowed Eni a share in the profit split on gas redirected from Italia to global LNG markets. Although Italy's gas consumption shrinks to 70 billion m³ this year from 73.4 billion m³ in 2012, competition is meanwhile growing, as will supply diversity once Azeri gas imports start in 2019. That makes it unlikely that the proposed 8 billion m³/year Galsi pipeline linked Algeria to the Italian mainland via Sardinia will progress beyond the concept stage.

Spot LNG prices in Northeast Asia rose up to 15.35USD/ MMBtu as most market players concluded October transactions, but slid in Southwest Europe to 11.50USD/ MMBtu. Netbacks for Middle East sellers were about 3.50USD/MMBtu higher on Asian than Southwest Europe LNG deliveries, while UK-Belgian netbacks for Middle East sellers lagged Asia by roughly 5.75USD (Table 3).

U.S. natural gas prices in domestic market are moderately recovering with the prompt contract currently trading at about 3.5 - 3.7USD/MMBtu. The recovery was attributed to storage increase and favourable weather expectations, including stirring tropics. However, the upside remains capped by continued gas production growth. Much attention has focused on how the Henry

| Netbacks in USD/MMBtu Exporters | Northeast Asia | Southwest Europe | UK | Belgium | U.S. Mid-Atlantic | U.S. Northeast | Mexico Altamira |
|------------------------------------|-------------------|---------------------|------|---------|----------------------|-------------------|--------------------|
| Qatar | 13.31 | 9.72 | 7.51 | 7.54 | 1.15 | 1.28 | 0.67 |
| Oman | 13.48 | 9.87 | 7.77 | 7.67 | 1.29 | 1.42 | 0.96 |
| Abu Dhabi | 13.35 | 9.76 | 7.59 | 7.58 | 1.19 | 1.40 | 0.76 |
| Indonesia | 14.36 | 8.94 | 6.89 | 6.68 | 0.57 | 0.66 | 0.44 |
| Malaysia | 14.30 | 9.01 | 6.97 | 6.99 | 0.55 | 0.65 | 0.65 |
| Australia | 14.10 | 8.90 | 6.89 | 6.82 | 0.71 | 0.80 | 0.61 |
| Russia | 14.88 | 8.47 | 6.54 | 6.57 | 0.12 | 0.17 | ? |
| Trinidad | 11.33 | 10.45 | 8.28 | 8.27 | 2.67 | 2.69 | 2.59 |
| Algeria | 12.08 | 11.20 | 8.71 | 8.70 | 2.34 | 2.47 | 2.02 |
| Egypt | 12.53 | 10.84 | 8.41 | 8.44 | 2.08 | 2.21 | 1.80 |
| Nigeria | 12.06 | 10.39 | 8.17 | 8.16 | 2.01 | 2.10 | 1.84 |
| Equatorial Guinea | 12.15 | 10.46 | 8.21 | 8.23 | 2.09 | 2.17 | 1.88 |

Table 3. Spot LNG Exporter netbacks at key markets worldwide

Source: WGI, 11 September 2013

Hub cash market for natural gas will influence world gas markets when the U.S. begins exporting LNG. Based on a study released by PIRA Energy group in New York, the Henry Hub spot prices will become significantly more volatile once U.S. LNG exports begin, and the more export capacity that is approved and built, the greater the volatility will be. Price volatility at Henry Hub will be influenced by LNG-related changes in the supply and demand balances within the North American market. With U.S. LNG exports expected to crest at 9 billion ft³/ day by 2025, PIRA analysts said, "The call on U.S. gas production will account for 5 - 15% of the total. The timing of new supply, combined with domestic gas demand growth from low Henry Hub prices and the stability of LNG production, add significant layers of complexity. The changing dynamic in the market is that the fairly insular world of North American gas markets and Henry Hub pricing will be immediately exposed to supply, demand, inventory, and pricing issues in other parts of the world". A global network of buyers and sellers will integrate North American LNG supplies within existing portfolios and impose a completely new dynamic on North American gas markets. Foreign buyers will pay a premium to Henry Hub to acquire North American LNG, but they will seek a discount relative to their current primary sources as well. The open nature of the North American gas market is uniquely attractive to foreign buyers because it allows them unprecedented reach within the entire LNG value chain. This wide-ranging penetration combines with their engagement with other global LNG producers to herald a reconfiguration of the entire LNG business. Once LNG exports occur en masse in North America, the interaction between global gas prices and the Henry Hub price will be bi-directional. The simply rule of supply and demand dictates that. Some will win and some will lose, PIRA analysts said.

Parallel to USA, Russian gas export policy's shift to LNG matters. Russia is changing its natural gas export emphasis from dry gas through westbound steel pipelines to liquefied natural gas aboard tankers bound for East Asian markets and South Korea, Thane Gustafson, a Georgetown University professor said. Gazprom adapted in some ways the last 30 years by taking downstream equity positions in European pipelines and distribution systems as far away as Great Britain. But it is facing resistance to new pipeline systems as Europe begins to diversify its sources and countries there consider importing LNG. Meanwhile, Russia's domestic gas market, which Gazprom executives long considered a graveyard because they had to sell gas

nearly for free over several decades, has started to come to life as a market economy has grown and prices have risen. Gazprom, which originally supplied 90% of Russia's domestic gas needs, has seen its share fall to 70% as competition from independent producers has grown. It also was slow to realise LNG's potential as it developed supplies in the Yamal peninsula, giving it enough dry gas for the foreseeable future at a time when other producers globally turned their attention to shale gas. Gazprom finally developed a maritime transportation unit with tankers that carry other countries' LNG to customers in Europe and North America. Gazprom Export and South Stream Transport B.V. also signed a gas transmission agreement at the October 2 meeting, Gazprom said. It is constructing the pipeline to ship gas across the Black Sea to Central and Southern Europe. To the East direction, Gazprom and China National Petroleum Corp (CNPC) signed an agreement covering major terms and conditions of supplying natural gas from Russia to China via the eastern route, in accordance with terms previously reached. Earlier negotiations covered the supply of 30 billion m³/years starting in 2015. Pricing will not be linked to the Henry Hub.

According to the most recent World Energy Outlook of the IEA, Southeast Asia's growing energy consumption is leading to the region's increasing reliance on oil imports, imposing high financial costs and external vulnerability on the region's economies. "Southeast Asia is, along with China and India, shifting the centre of gravity of the global energy system to Asia", IEA Executive Director Maria van der Hoeven said. Southeast Asia's energy demand will increase by more than 80% in the period to 2035, a rise equivalent to current demand in Japan. By 2035, the region is projected to become the world's fourth-largest oil importer after China, India, and the European Union, with its oil imports reaching more than 5 million b/d and its annual spending on oil imports rising to 240 billion USD, accounting for 4% its GDP. Its oil dependency will so be doubled to 75% of demand. Southeast Asia's surplus of natural gas and coal exports also will decline as production is increasingly diverted to domestic markets. The region's net gas exports decline to 14 million m³ in 2035. Serious actions should be taken by countries in the region to improve energy efficiency. IEA also highlighted that developing policies to attract investment is vital for enhancing energy security, affordability and sustainability in Southeast Asia. In the period to 2035, around 1.7 trillion USD of investment in energy- supply infrastructure is required.

LNG prices in Northeast Asia jumped 20 cents to 15.55USD/MMBtu as buyers opened talks on early-November deals ahead of winter, but held steady at 11.50USD/MMBtu in Southwest Europe. Asian prices are now on the rise again to mid-16USDs from October to November 2013. Potential sources include Nigeria, Peru, Yemen, Trinidad & Tobago, and perhaps a few Atlantic reloads with high shipping costs. Spot availability is unclear from traditional Pacific producers.

| | Current | Previous |
|----------------------|---------|----------|
| Northeast Asia LNG | 15.55 | 15.35 |
| Southwest Europe LNG | 11.50 | 11.50 |
| | Spot | Futures |
| Britain' s NBP | 10.64 | 10.56 |
| U.S. Henry Hub | 3.63 | 3.74 |

Table 4. Indicative LNG, Spot gas prices (USD/MMBtu)

Source: WGI, 18 September 2013

Oil and gas pipeline construction markets

From 2003 to 2007 U.S. oil and gas pipeline operating revenues increased continuously from 15,08 to 21,73 billion USD, but in the next two years, they came down to 18,95 billion USD in 2009. After that, from the end of 2009 to present day, theirs rising trend is repeated with a rate lesser comparing to the first period (Table 5a).

Oil pipeline operators' net income continued to rise from 2003 to 2013 with some variations in short cycles. The resulting earnings as a percent of revenue eased from the record levels seen in 2011 to 48,64% (Table 5b). Natural gas pipeline companies in 2012 saw operating revenues rise by more than 424 million USD or roughly 2.1% from 2011, continuing the gains seen the year before. Net incomes, however, fell roughly 123 million USD (more than 2.5%), also continuing the trend begun in 2010 (Table 5a). U.S. pipeline operators used their rising revenues and net incomes to grow their systems and expanded plans for future growth. Oil pipeline investment components are as follows:

- For crude oil: Line pipe and fittings 25.26%; Land and right-of-way (ROW) 4.25%; Misc. 4.03%; Pumping station and equipment 20.43%; Pipeline construction 46.02%.

- For products: Line pipe and fittings 23,54%; Land and ROW 2.86%; Misc. 7.81%; Pumping station and equipment 30.85%; Pipeline construction 34.95%.

- Pipeline construction costs - estimated: Materials 23.20%; ROW and damages 2.58%; Labour 38,84%; Misc. 35.37%.

The land pipeline construction costs were 4.1 million USD/mile. Pipeline labour prices remained the single most expensive per-mile item at 1.6 million USD/mile, but the biggest single increase was seen in material costs, which nearly doubled to 952,328USD/mile. Estimated land pipeline construction costs for projects completed in the 12 months ending June 2013 were nearly 600,000USD/ mile higher than actual costs, more than 400,000USD/ mile of which came from inflated miscellaneous cost estimates. Actual compressor station costs were 5.1% less than estimated costs for projects completed by June 2013, with significantly smaller miscellaneous costs here as well cancelling out much higher than anticipated labour cost.

The estimated costs for new offshore projects in 2012 - 2013 were 7.6 million USD/mile.

Variations over time in the four major categories of pipeline construction costs - material, labour, miscellaneous, and ROW - can also suggest trends within each group.

| Year | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | 2009 | 2010 | 2011 | 2012 |
|----------------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Operating revenues | 15.08 | 15.78 | 16.37 | 17.12 | 21.73 | 19.80 | 18.95 | 19.79 | 20.55 | 20.97 |
| Net incomes | 3.26 | 3.59 | 3.86 | 4.02 | 4.76 | 5.10 | 4.65 | 5.21 | 4.89 | 4.76 |
| Net inc./Oper.revenues (%) | 21.62 | 22.75 | 23.58 | 23.48 | 21.90 | 25.75 | 24.54 | 26.32 | 23.79 | 22.70 |

b) Oil pipeline

a) Gas pipeline

| Year | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | 2009 | 2010 | 2011 | 2012 |
|----------------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Operating revenues | 7.70 | 8.02 | 7.92 | 8.52 | 8.99 | 9.24 | 9.98 | 11.22 | 12.56 | 14.01 |
| Net incomes | 3.47 | 3.32 | 3.07 | 3.74 | 3.75 | 3.93 | 4.13 | 4.58 | 6.11 | 6.42 |
| Net inc./Oper.revenues (%) | 45.06 | 41.39 | 38.76 | 43.89 | 41.71 | 42.53 | 41.38 | 40.82 | 48.64 | 45.82 |

Source: Oil & Gas Journal, 2 September 2013

| Categories | Length (miles) | Material | Labour | Misc. | ROW and Damages | Total |
|-----------------------------------|-------------------|----------|-----------|-----------|--------------------|-----------|
| Total projects - land | 820.12 | 781.023 | 1,307.536 | 1,190.626 | 86.894 | 3,366.079 |
| Total land - 2012 report | 144.05 | 71.423 | 199.261 | 155.598 | 20.373 | 446.655 |
| Total projects - offshore | 22.90 | 33.959 | 89.963 | 51.022 | 0 | 174.944 |
| Total - all projects | 843.02 | 814.981 | 1,397.499 | 1,241.648 | 86.894 | 3,541.023 |
| 2012 - report total, all projects | 144.05 | 71.423 | 199.261 | 155.598 | 20.373 | 446.655 |

Table 6. U.S. pipeline costs, estimated (million USD)

Source: Oil & Gas Journal, 2 September 2013

| Table 7, 2013's U.S. Land | nineline construction costs | (average cost_USD/mile) |
|----------------------------------|-----------------------------|--------------------------|
| INDIC 7. 2013 5 0.3. Lunu | | (average cost, osb/minc) |

| Pipeline size | ROW | Material | Labour | Misc. | Total |
|---------------|---------|-----------|-----------|-----------|-----------|
| 8 in. | 71,443 | 188,261 | 69,541 | 1,533,654 | 1,862,899 |
| 12 in. | 64,313 | 319,004 | 784,464 | 380,252 | 1,548,033 |
| 16 in. | 81,810 | 286,739 | 533,749 | 636,324 | 1,538,623 |
| 20 in. | 103,333 | 338,025 | 998,560 | 701,317 | 2,141,235 |
| 24 in. | 73,560 | 623,116 | 805,886 | 912,622 | 2,415,184 |
| 36 in. | 93,529 | 1,400,946 | 2,182,912 | 1,938,652 | 5,616,040 |

Source: Oil & Gas Journal, 2 September 2013

Materials can include line pipe, pipe coating and cathodic protection. "Miscellaneous" costs generally cover surveying, engineering, supervision, contingencies, telecommunications equipment, freight, taxes, allowances for funds used during construction (AFUDC), administration and overheads, and regulatory filing fees. ROW costs include obtaining ROW and allowing for damages. For the 26 land spreads filed for in 2012 -2013, cost-per-mile projections rose in all categories except ROW and damages, which eased for the third straight year. In 2011 miscellaneous charges actually passed material to become the second most expensive cost category and they retained this position through

2013: Material - 952,328USD/mile, up from 495,821USD/ mile for 2011 - 2012; Labour - 1,594,323USD/mile, up from 1,383,277USD/mile for 2011 - 2012; Miscellaneous -1,451,770USD/mile, up from 1,080,169 USD/mile for 2011 - 2012 (the continued rise in miscellaneous costs was driven by companies increasing the amount set aside for contingencies in their estimates); ROW and damages -105,953USD/mile, down from 141,431USD/mile for 2011 - 2012. The average cost per mile for any given pipeline diameter may fluctuate year to year as projects' costs are affected by geographic location, terrain, population density or other factors. **Ha Phong**

