# 科学研究費助成事業

研究成果報告書



令和 3 年 6 月 1 4 日現在

機関番号: 17102
研究種目:若手研究
研究期間: 2019 ~ 2020
課題番号: 19K15490
研究課題名(和文)Mitigating Asphaltene Deposition Using Modified Poly-Vinyl Alcohol and Silica Nanoparticles
研究課題名(英文)Mitigating Asphaltene Deposition Using Modified Polyvinyl Alcohol and Silica Nanoparticles
研究代表者
NGUELE RONALD (Nguele, Ronald)
九州大学・工学研究院・助教
研究者番号:50821401
交付決定額(研究期間全体):(直接経費) 1.700.000円

研究成果の概要(和文): The research investigated the synergy between silica oxide nanoparticles and polyvinyl alcohol in mitigating the deposition of the deposition in asphaltenes, which is a major challenge in oil and gas industry.

#### 研究成果の学術的意義や社会的意義

the present research proposed a hybrid production scheme in which the nanofluid should be injected ahead of the carbon dioxide. This approach promotes the adsorption the polar asphaltenes onto the surface of the nanoparticles allowing thereby oil dissolution by CO2.

研究成果の概要(英文): It was found that asphaltenes, extracted from dead crude oils develop a stronger affinity with the silica nanofluid film leading to a preferential adsorption. Also, the research showed that the adsorption is proportional with the aromatic core, and inversely proportional to heteroatom contents. It was further highlighted that asphaltene solubility is rather dependent of the size of the aromatic core and the amount of embedded heteroatoms.

研究分野: Petroleum Engineering

キーワード: CO2-EOR Asphaltene Nanofluid Poly vinyl alcohol

科研費による研究は、研究者の自覚と責任において実施するものです。そのため、研究の実施や研究成果の公表等については、国の要請等に基づくものではなく、その研究成果に関する見解や責任は、研究者個人に帰属します。

# 1. 研究開始当初の背景

In general, enhanced oil recovery (EOR) is the last stage of oil production, which is implemented when both the primary (oil produced by natural formation drive) and the secondary (injection of water and/or gas) stages become economically not attractive. Most common EOR schemes are classified based on the material/agent used to enhance the production. They are either water-, chemical- and heat- or gas-based; each of aforementioned techniques has sub-sets.

Gas-based EOR is particularly attractive because it offers the possibility to sequester CO<sub>2</sub>, whose concentration accounts for about 99% of the overall emitted gas. EOR, which has the further potential to displace stranded oil. Even if the current market reports that up to 40% of the residual oil could be extracted from a depleted oil reservoir using gas injection, CO<sub>2</sub>-EOR has not yet reached an acceptable exposure. The reason behind this drawback is that CO<sub>2</sub>-EOR is associated with the well-documented organic deposition.

Organic deposition is a general term to describe the formation of precipitates that consist primarily of asphaltenes, wax and resins. The former is in suspension in the crude oil and precipitate after the aggregation, crystallization of the suspended particles. Because these aforementioned mechanisms start at the nano-level, it is reasonable to think that a material that could reduce (or at least control) the impingement of asphaltenes could delay the precipitation and thus enhance the oil production.

With this belief in mind, nanomaterial appears as possible candidate. NC is defined as a material made from two or more components with significant different physical and/or chemical properties with a least one of the individual phase being within the range of nanometer (1nm=10-9 m). At a nano-level, individual properties of a material are tremendously enhanced, among which adsorption, which is so because the ratio surface-to-volume is increased.

Because of these features, the present research proposes to inhibit in-situ the aggregation of asphaltenes, following an injection in which  $CO_2$  will be injected at the trail of a stabilized NP. This approach, in theory, promote the adsorption the polar asphaltenes onto the surface of the NC allowing thereby oil dissolution by  $CO_2$  (**Fig. 1**).



### Fig. 1 Proposed research concept

### 2. 研究の目的

The primary goal of this research is to improve oil production using  $CO_2$  gas injection. The subset objectives deriving therefrom are (1) the characterization of asphaltenes; (2) the evaluation of Interactions asphaltenes/NPs – IFT.

### 3.研究の方法

Asphaltene sample, extracted from a dead heavy oil using n-heptane using standard

procedures. The spectral characterization of asphaltenes included Energy-dispersive X-ray spectroscopy (EDX), Raman spectroscopy as well as mass spectrometry. The mitigation was performed on modeled crude oil consisting of extracted asphaltene dissolved in pure toluene. The silica-based nanofluid was prepared by dispersing silica oxide nanoparticle (0.1 wt%) into an aqueous solution of modified polyvinyl alcohol (1 wt%). Adsorptions tests were conducted on Berea sandstone in the presence of carbon dioxide (CO2) bubbling.

# 4. 研究成果

Four n-heptane asphaltenes were precipitated from oilfields located in Northern Asia, West Africa, Middle East, were investigated. Computed from D1 and G bands of the Raman spectra, the aromatic diameter sheet ( $L_a$ ) was found between the range of 0.8 to 1.8 nm. The analysis of the mass spectrograms revealed that the molecular weight (MW) ranges from 273 to 345 Da (**Fig. 2**).



Fig 2 Relative asphaltene solvency in different solvents

Also, it was found that asphaltenes develop a stronger affinity with the silica nanofluid film leading to a preferential adsorption. Also, the research showed that the adsorption is proportional with the aromatic core, and inversely proportional to heteroatom contents. It was further highlighted that asphaltene solubility is rather dependent of the size of the aromatic core and the amount of embedded heteroatoms (**Fig. 3**).



(a) Asphaltene adsorption affinity



Fig. 1 Relationship between asphaltene desorption, adsorption affinity and asphaltene selfassociation

## 5.主な発表論文等

# 〔雑誌論文〕 計1件(うち査読付論文 0件/うち国際共著 0件/うちオープンアクセス 0件)

1.著者名	4.巻
Nguele Ronald, Sasaki Kyuro	622
2.論文標題	5 . 発行年
Asphaltene behavior at the interface oil-nanofluids: Implications to adsorption	2021年
3. 雑誌名	6.最初と最後の頁
Colloids and Surfaces A: Physicochemical and Engineering Aspects	126630 ~ 126630
掲載論文のDOI(デジタルオブジェクト識別子)	査読の有無
10.1016/j.colsurfa.2021.126630	無
オープンアクセス	国際共著
オープンアクセスではない、又はオープンアクセスが困難	-

## 〔学会発表〕 計0件

〔図書〕 計0件

〔産業財産権〕

〔その他〕

6 . 研究組織

\_

_			
	氏名 (ローマ字氏名) (研究者番号)	所属研究機関・部局・職 (機関番号)	備考

## 7.科研費を使用して開催した国際研究集会

〔国際研究集会〕 計0件

# 8.本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関
---------	---------