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Research Article

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Direct Visualization of Ultrasonication Induced Asphaltenes Removal in Carbonate Rock Using Confocal Imaging

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Abstract

Asphaltenes deposition is a major issue in the petroleum industry as it can have a detrimental impact on hydrocarbon recovery efficiency. Therefore, it is imperative to study the fundamental mechanisms controlling the asphaltenes flocculation and deposition in reservoirs allowing us to prevent and possibly eliminate such problem. Hitherto many studies have highlighted ultrasonication as a potential remediation technique but no investigation has been able to provide direct visual evidence of the phenomena. The primary objective of this study is to visualize the deposition of asphaltenes and their subsequent removal by ultrasonication in Indiana Limestone using state of the art confocal microscopy. To do so, we performed a comprehensive series of experiments by flooding Indiana Limestone core samples with crude oil and later passing ultrasonic waves through the flooded sample. Four core samples of Indiana Limestone each displaying different permeability were used, these are referred to as A2, B2, C4, and D4. At each stage of experiment series of images were captured by confocal microscopy depicting asphaltenes deposition and it's post-sonication distribution. The images were further segmented allowing us to compute changes in the asphaltenes content before and after sonication. The comparison of confocal scans reveals that the ultrasonic irradiation is highly efficient in removing asphaltenes from the low permeability core samples, whereas in the case of highly permeable cores, rather than preventing it promoted the asphaltenes flocculation. Surprisingly, an increase in asphaltenes content was observed after ultrasonication in high permeability core samples.

Introduction

One of the important issues that challenge the oil and gas industry is the flow assurance problem caused by asphaltenes deposition [1]. Asphaltenes are large, complex and polar molecules in petroleum crudes. By definition, asphaltenes are compounds soluble in aromatic hydrocarbons such as toluene orxylene but are insoluble in alkane liquids such as n-heptane of crude oil. Asphaltenes take the form of suspended solids in the crude oil or can deposit onto the rock. This process eventually reduces the ability of the fluid to flow by clogging the pore space and altering its wettability and therefore effecting relative permeability and capillary pressure [2-5]. This plugging results in reduced productivity and increased costs.

There are many techniques used in the field to mitigate the issue of asphaltenes plugging. The technique employed in this study involves an alternative to existing technologies and suggests the application of ultrasonication to remove the deposition in the near-wellbore region. The technology involves the introduction of ultrasonic irradiation from a sonotrode passing through the core sample and generation of a vibration pattern that sets the asphaltenes deposited in the rock into motion. The ultrasonic vibration patterns cause the deposition in the rock to stretch, compress and relax.

The application of acoustic waves in removing asphaltenes deposits in porous media has been extensively studied. Gunel and Islam studied the role of ultrasonic irradiation on crude oil properties alteration. The study resulted in a change in the rheological properties of oil samples by the application of ultrasonic waves. Dunn and Yen demonstrated the effect of ultrasonic waves on the conversion of asphaltene molecules and found out that ultrasonication caused dehydrogenation and cracking in bitumen. Champion et al [6-8]. conducted a study where theyproduced high power sound along with a high voltage electrical discharge to explore the use of acoustic waves for wellbore cleaning. They found that high power sound wave technology is an effective method of removing asphaltene that causes wellbore plugging. Sohrabi et al [9]. performed micro-model experiment under pressure in order to investigate the effect of the ultrasound on production index of condensate gas reservoirs. The result showed that a significant amount of condensate gas with ultrasonic irradiation was produced [6]. In a recent findings Najafi et al [10]. presented possible benefits for the application of ultrasonic wave technology as a method of asphaltenes flocculation/deposition inhibition. An optimum radiation time at which ultrasonication had the most noticeable effects was reported. Another study conducted by Amani et al [11]. showed the effect of ultrasonic waves on asphaltenes precipitation in core samples. In their study, a core sample was flooded with heptane and crude oil and permeability measurement before and after sonication was conducted. They found out permeability increases after the application of ultrasonication on the core sample.

As shown in the previous paragraph, that many previous studies have demonstrated the ability of ultrasonication in preventing asphaltenes flocculation as well as unplugging the pore space initially clogged by asphaltenes precipitates. However, these studies cannot be considered conclusive as they relied on macroscopic properties such as a change in permeability of core samples to test the effectiveness of ultrasonication. Therefore, to date, according to our knowledge no direct visual evidence is reported in the literature that can confirm the exact effect of ultrasonication on asphaltenes deposits. Though the recent study by Dehshibi et al [12]. have been able to delineate the ultrasonic irradiation-induced oil recovery via pore-scale images, the experiments were conducted in 2D micromodel with simplified pore space representation.

In this research, we leverage the confocal laser scanning microscope technology CLSM to evaluate the precipitation as well as the removal of asphaltenes via ultrasonication in real carbonates rocks, providing direct visual evidence of control of ultrasonic waves over asphaltenes precipitates. Overall, our research aids in improving the understanding of asphaltenes kinetics at the fundamental level and can play an integral role in modeling the phenomena at the macroscopic scale.

Experimental Methodology

In this section we describe the experimental setup used to understand the deposition and remediation using ultrasonication. The general idea for this section of the study is to provide a visual representation by using confocal laser scanning microscope technology, CLSM. Series of experiments were performed using different samples of Indiana Limestone A2, B2, C4 and D4. The length of core sample was kept constant at 5 cm and dimeter at 2.5 cm. The properties of these core samples are listed in Table 1.

Table 1: Showing properties of core samples used for the experiments.

Core Sample	Porosity (%)	Permeability (mD)
A2	8.25	13.5
B2	8.41	1.10
C4	6.32	49.80
D4	6.51	178.0

It can be seen from Table 1 while the porosity values of core samples are almost similar, there is significant variation in their permeability values. Three stages of experiments were planned and carried out accordingly. These stages are laid out in Figure. 1.

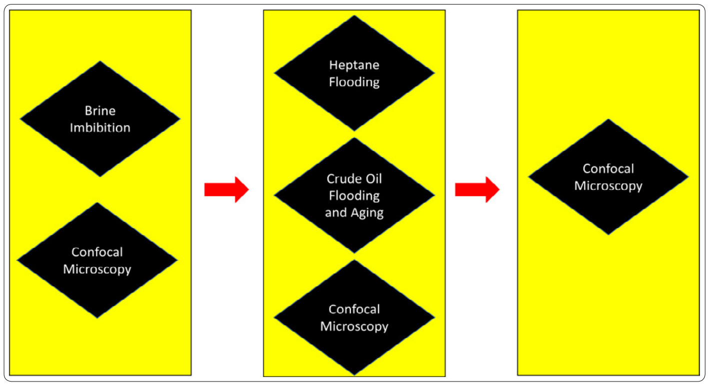

Figure 1: Various stages of experiments.

During Stage 1, the core sample was prepared. This involved saturating the core sample with brine (Figure. 2), and then confocal scan were obtained to serve as baseline data (see Figure. 3 for the confocal microscopy setup).

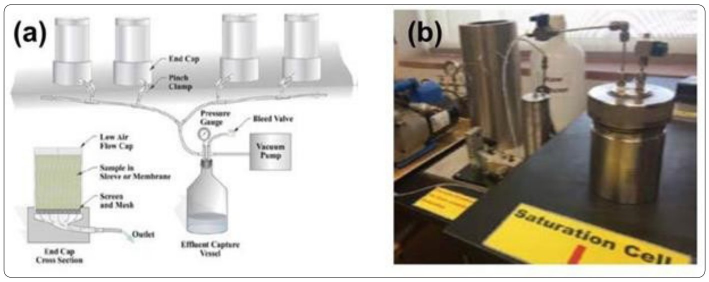

Figure 2: (a) Schematic of brine Saturation Setup. (b) real experimental setup for brine saturation in the lab.

Figure 3: Leica TCS SP8 Confocal Microscope Setup.

Stage 2 commenced with core flooding using light hydrocarbon (heptane) and the crude oil. The flooded sample then was aged overnight. This process was done for asphaltenes precipitation in the core sample. Later, confocal microscopy scans were conducted to serve as data before undergoing remediation. After conducting the tests of Stage 2, the core sample was subjected to ultrasonic wave for 7.5 minutes (optimized timed as found by Najafi and Amani at 90% amplitude using the UIP1500hd from Hielscher Ultrasound Technology (Figure. 4). Confocal scanning was then conducted. These final tests were held in comparison with the initial result before remediation [13].

Figure 4: Showing Ultrasound apparatus used in the experiments.

All confocal scanning was done with the Leica TCS SP8 Confocal Microscope (Figure. 3) [14]. Initially, two settings were executed, namely reflection and fluorescence modes. Fluorescence mode responds only to materials that when subjected to energy of certain wavelength, excite and give off signal that can then be collected and processed to form images. On the other hand, reflection mode operates when energy is released into the sample and gives back signal to the processor as it hits any material along its path. In this project, reflection mode was used since the images it gave are clearer than the ones from fluorescence mode.

We used the 10X lens with a Numerical Aperture of 0.30. The pixel size of all images is x/y = mani284nm, z=4.2µm with pixel dimensions x/y 4096 X 4096 having Z as varied. The fluorescence images were executed using a 405nm laser and the emission was collected from 450 nm to 580nm. The reflection mode images were imaged with a 488nm laser light source, and the reflection was of 450 to 580 nm window.

Results and Discussion

Asphaltenes, can absorb light well and give strong fluorescence and therefore are clearly visible as persistent patches of white coloration as shown in Figure. 5 (a). It is important to note that during the confocal microscope scan, the exact location for the scans cannot be practically achieved due to the fine resolution of the images [15].

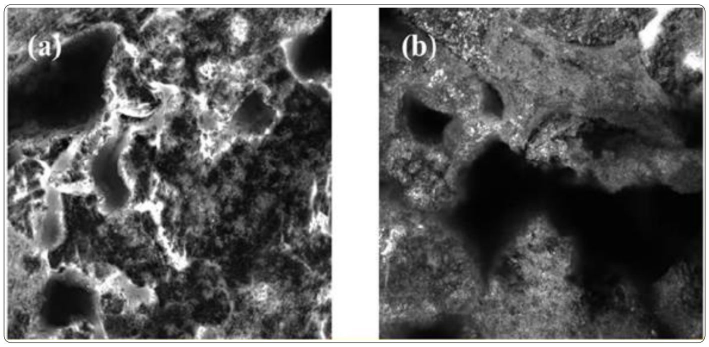

Figure 5: State of core sample A2 (a) before and (b) after sonication. The intense white color shows the asphaltenes deposits**.**

Comparison of A2 core sample before (Figure. 5 (a) and after sonication (Figure. 5 (b) indicates a decrease in the intensity of white coloration after passing ultrasonic waves through the sample manifesting reduction in asphaltenes content. As the core sample is exposed to the ultrasonic irridation, the asphaltenes deposits get highly unstable. The ultrasonic waves engender a perturbing environment that exerts stress on asphaltenes deposits and eventually disintegrating them into smaller particles increasing their surface area. The disintegrated asphaltenes particles either get washed away out of core sample or get dissolved into crude oil [13,16,17]. Overall, Figure. 5 shows the remarkable impact of sonication on asphaltenes precipitates and confirms the existing literature that suggests sonication as an effective remediation technique for reducing asphaltenes deposits and restoring the fluid conducting capacity.

The condition of core samples A2, B2, C4, and D4 before and after sonication can be seen in Figure.6.

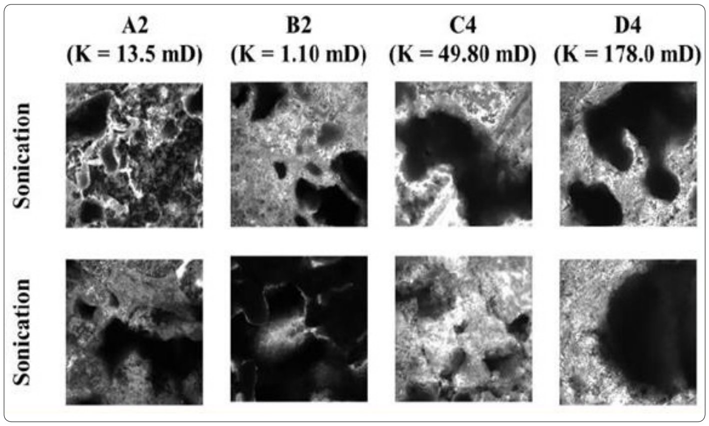

Figure 6: Images of core sample before and after sonication.

Close inspection of Figure. 6 indicates that the permeability of core sample may control the asphaltenes removal via ultrasonication. There is a significant change in the white coloration of the low permeability core sample A2 and B2, however, apparently, the intensity of white coloration seems to remain almost similar before and after sonication in high permeability core samples C4 and D4. This further illustrates that the efficiency of ultrasonication as a remediation technique in porous media may inherently depend on the morphological and topological characteristics of pore space. However, to confirm this further investigation is required this is our future objective.

To quantify the asphaltens content in the investigated core samples we segmented the confocal images using ImageJ version 1.52p, the details of segmentation procedure are given in Figure. 7.

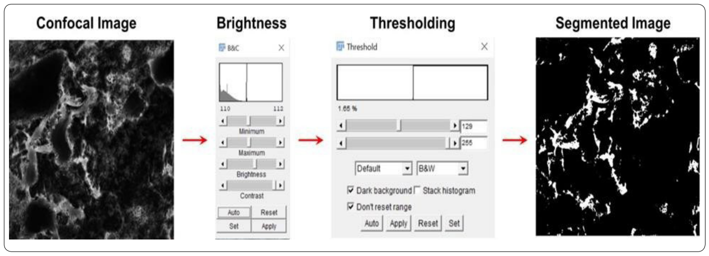

Figure 7: Segmentation procedure for confocal images**.**

Segmented images for each of the core samples are shown in Figure.8.

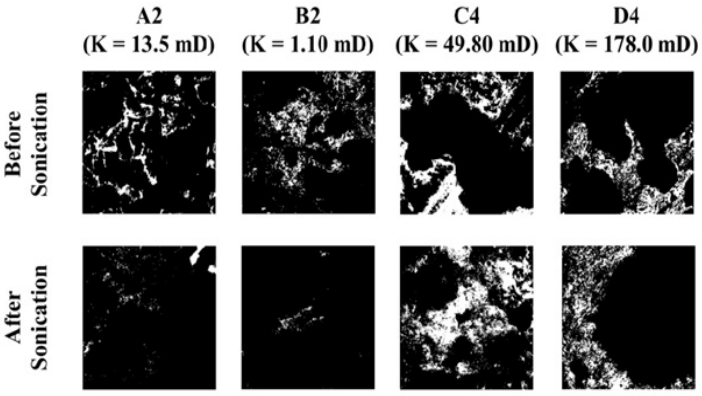

Figure 8: Segmented images of core sample before and after sonication. The white color represents the asphaltenes deposits**.**

Using segmented images the asphaltenes content before ultrasonication process Db and after ultrasonication process Da was quantified for all the core samples. Figure.9 (a) manifests parabolic relationship between permeability and Db. The amount of asphaltenes precipitated in porous media increases as the permeability of porous media increases, until it reaches a threshold value after which Db is inversely related to the permeability of core sample. Moreover, the trend shown for Da against permeability in Figure. 9 (b) is similar to Figure. 9 (a). We also presented the comparison of Db and Da in form of a histogram is shown in Figure. 10.

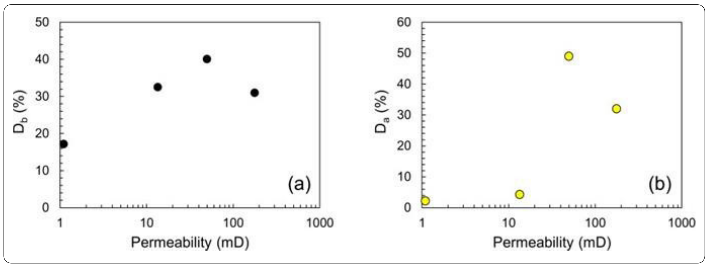

Figure 9: (a) Db as a function of permeability. (b) the non-monotonic relationship between Da and permeability.

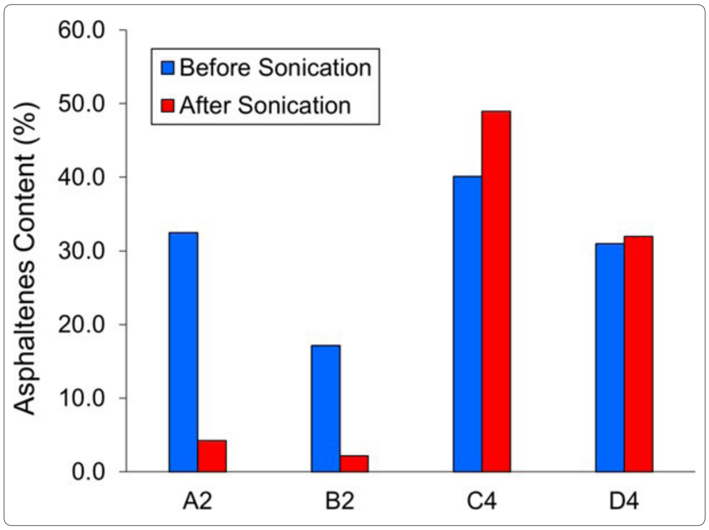

Figure 10: Histogram illustrating the comparison between asphaltene content before ultrasonication Db and after ultrasonication Da.

It clearly demonstrates that though the asphaltenes content decrease after sonication in core sample A2 and B2, surprisingly in the case of C4 and D4 the extent of asphaltenes precipitates increases after sonication. It is important to note that A2 and B2 are tight (low permeability) carbonate system, while C4 and D4 represents high permeability rock sample. It has been shown in the investigation by Najafi and Amani that asphaltenes kinetics is a strong function of ultrasonic radiation time Rt, which is parabolic in nature. According to Najafi and Amani there is an optimum radiation time $R*$ before which the asphaltenes content decreases with Rt and after which asphaltenes content increases with Rt. As stated by Najafi and Amani before $R*$ asphaltenes particles disintegration is the dominant process, and after $R*$ amalgamation of free radical to form new asphaltenes plays a crucial role in the precipitation of asphaltenes particles [13].

Using the theory proposed in Najafi and Amani of $R*$ we can explain the results presented in Fig. 10. In this study, the core samples were exposed to ultrasonic radiations for 7.5 mins. In the case of A2 and B2 (low permeability core samples) the $R*$ may be greater than 7.5 mins, while in the case of C4 and D4 (high permeability core sample) the $R*$ may be well below 7.5 mins. As a consequence, a decrease in the asphaltenes deposits is noticed after sonication in core ample A2 and B2, whereas in the case of

C4 and D4 the effect of ultrasonication on asphaltenes kinetics is reversed (Da > Db).

We further show a phase diagram (Figure. 11) that distinguishes two regions shaded blue and yellow The blue region depicts efficiency of ultrasonication Db - Da > 0, this region may represents condition when $Rt < R*$ therefore the asphaltenes particles disintegrates and subsequently are washed away out of the core sample. Whereas the yellow region shows negative efficiency Db - Da < 0 as a result of $Rt > R*$.

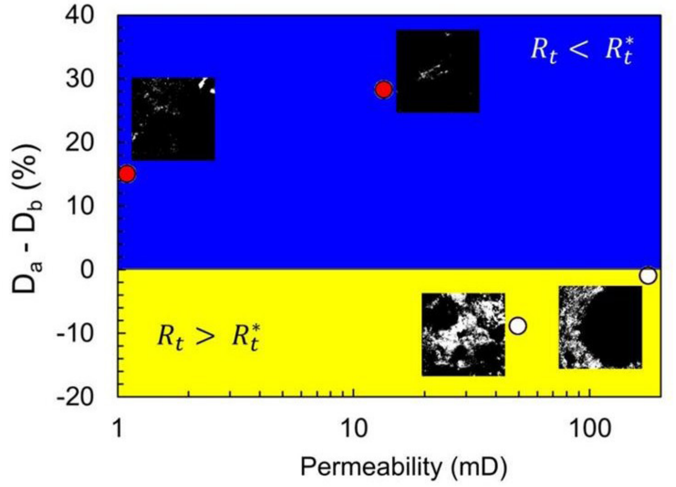

Figure 11: Phase diagram that differentiates $Rt < R*$ region shaded blue and $R < R*$ region shaded yellow. The fille points represents Db - Da > 0 and empty points shows Db - Da < 0.

Conclusion

In this research comprehensive series of experiments were performed to observe the deposition of asphaltenes in Indiana limestone core samples and the effectiveness of ultrasound irradiation as aremediation technique using the confocal laser scanning microscopy technology. Our study provides first-ever direct visual evidence of asphaltenes removal via ultrasonication in real rock samples confirming the results reported previously in the literature. Moreover, the results demonstrate that the permeability of porous media may have a significant influence on the asphaltenes kinetics. We hypothesized that the optimum ultrasonication radiation time will vary depending upon the permeability of core sample, which will impact efficiency as well as the role of ultrasonication. Our future goal is to conduct experiments to further determine the control of permeability of core samples on the efficiency of ultrasonication.

Acknowledgment

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