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Investigating the effects of ultrasonic waves and nanosilica on the viscosity reduction of Sharqy Baghdad heavy crude oil

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Abstract

The current study examines the combined impacts of ultrasonic waves and nano silica (NS) on reducing the viscosity Sharqy Baghdad heavy crude oil with an API gravity of 20.32. NS of an average particle size of 59.93 nm and 563.23 m²/g surface area were produced utilizing the sol-gel technique from Iraqi sand. The XRD analysis indicates the existence of an amorphous silica, the SEM analysis showed that NS tends to agglomerate, and the FTIR spectra exhibited the presence of siloxane and silanol groups. In addition, the TGA analysis demonstrated a total weight loss of 15.62%, validating the thermal stability of the NS. The experiments included a study of the impact of ultrasonic power, exposure time, duty cycle, temperature, and the combined effects of the ultrasonic waves and silica nanoparticles on the degree of viscosity reduction percentage (DVR%). The results demonstrated that the viscosity of the heavy crude oil decreased by 27.83% at an irradiation time of 2 min, power of 360 W, 0.8 duty cycle, a temperature of 35 ºC, assisted by nano silica at a concentration of 1500 mg/L.

Keywords: Asphaltene; Heavy crude oil; Viscosity reduction; Ultrasonic waves; and Nanosilica.

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1- Introduction

Crude oil is a wide variety of naturally occurring, non-uniform compounds. It is a compound consisting of hydrocarbons with different structures, saturation levels, and various impurities [1]. The continuing worldwide demand for petroleum-based fuels has led to a decrease in production from conventional reservoirs, leading to a subsequent reaction in the markets, increasing oil prices, which in turn stimulated the development of techniques for the use of unconventional oil reservoirs in the global oil industry [2]. The increasing decline of conventional crude oil has led to a growing interest in the enhancement of heavy oil, which is widely recognized as a significant energy resource [3, 4].

Heavy oils are characterized by their elevated concentrations of hetero-compounds, including metals, sulfur, nitrogen, and oxygen, alongside a significant fraction of high-molecular-weight hydrocarbons [5,6]. Transportation of oil has become a complex and technologically advanced procedure, which necessitates the implementation of efficient and cost-effective methods for handling high-viscosity fluids. Due to its high density, which is comparable to, or in some cases, even surpasses, that of water [7], high sulfur content, and the presence of several metals, viscous crude oil cannot move through the pipeline. This poses numerous challenges throughout heavy oil production, separation, transportation, and refining [8]. The reason for these problems is asphaltene precipitation, which reduces the permeability of reservoir rock and changes the wettability of rocks, all of which have an unfavorable impact on the production from the reservoir rock [9]. Another possibility is the precipitation of asphaltene during the pipeline movement of oil. Additionally, asphaltene is the main contributor, leading to the elevated viscosity observed in heavy crude oils [10].

One significant issue associated with heavy crude oil is its tendency to produce a greater proportion of residual fraction and a lesser proportion of gasoline and diesel fuels during the distillation process. In addition, heavy crude oil needs to be refined using advanced techniques because it has a higher impact on the environment than its lighter equivalent [11]. Therefore, the main challenge is to reduce the viscosity of heavy oil, as this reduces internal friction resistance in the oil and enhances its mobility. So, viscosity reduction is very important for heavy oil production, pumping, and transportation [12]. There are various methods for improving heavy crude oil. However, current conventional techniques, both with and without using catalysts, often require high temperatures, longer reaction times, high prices, and seriously harm the environment. These methods include heating, dilution, emulsification, and core annular flow [13]. As a result, several new (unconventional) technologies for improving heavy oil include the use of magnetic fields, ultrasonic energy, and electric fields [14, 15].

From a technological, ecological, and economical viewpoint, Ultrasonic treatment has been identified as the optimal method to control the rheological characteristics

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of heavy oils due to their ease of use, cost-effectiveness, and ability to mitigate reservoir contamination [16]. By breaking the heavy molecules, ultrasonic can enhance heavy oil feedstock by producing cavitation bubbles [17]. Cavitation, which occurs when gas bubbles within the working fluid collapse can cause an increase in temperature and pressure [18].

Abramov et al. [19] have revealed that while oil production increased by 26.5%, viscosity decreased by 16% 4 hours after ultrasonic treatment, and the speed at which the oil rose from the well increased. Najafi et al. [20] studied the effective use of ultrasonic wave technology to solve the issues with asphaltene precipitation in Iranian oilfield. They found that 10 to 20 minutes of irradiation is the optimal time at which crude oils have the least viscosity and flocculation, and ultrasonic treatment can decrease the potential for the production of macrostructure aggregates. Kadyirov and Karaeva [16] examine the impact of ultrasound waves on the rheological properties of crude oil using two samples of 29.1 and 15.9 API gravity. They found that the viscosity decreased through ultrasound treatment, which reduces the cost of manufacturing and transportation. They found that the second sample exhibited a greater reduction in viscosity than the first sample.

The field of nanoparticle technology has grown rapidly and holds potential as a viable alternative to current methodologies employed for the upgrading and recovery of heavy oil [21]. Nanoparticles have specific characteristics that make them highly desirable as sorbents. In contrast to particles in bulk form, these particles exhibit significantly greater surface areas relative to their mass [22]. The nanoparticles have thermal catalytic capabilities that facilitate the efficient suspension of asphaltene within the oil, hence preventing its precipitation [23].

According to Montes et al. [24] they highlighted the potential of nanoparticles to decrease viscosity in heavy oils through two mechanisms: the disruption of the viscoelastic network and the catalytic degradation of the heavy portion following ultrasound treatment. The ultrasound cavitation process was examined using emulsified heavy oil of 13º API with 40 vol.% water during various exposure periods and SiNi1 nanoparticle dosages. A reduction in viscosity and asphaltene content was 44 and 16%, respectively.

The objective of this study is to investigate the decrease in viscosity of Sharqy Baghdad heavy crude oil by ultrasonic treatment using different ultrasonic parameters: power, irradiation time, duty cycle, and temperature. Furthermore, the use of prepared nano silica as a means of improving the efficiency of ultrasonic waves in decreasing the viscosity of heavy crude oil.

2- Experimental work

2.1. Materials and chemicals

The crude oil sample chosen for the present research was obtained from the Sharqy Baghdad oil field. Table 1 presents the characteristics of the heavy crude oil. The sand, which is a precursor for nanosilica, was sourced from the Local State Company for Mining Industries, Department of Mineral Extraction. The sodium hydroxide (NaOH) pellets, which were obtained from Riedel-de Haen AG, Seelze-Hannover, Germany, had a purity level of 99%. The malonic acid ($CH_2(COOH)_2$), with a purity of 98%, was provided by The British Drug Houses LTD./England. Distilled water was also used in the experiment.

Table 1. Heavy crude oil's characteristics

API° Gravity at 15.6 ºC	20.32
Density at 15.6 ºC, g/cm^3	0.932
Dynamic Viscosity at 40 ºC, cp	42.6
Asphaltene content, wt%	6.379
Sulfur content, wt%	4.68

2.2. Preparation of silica nanoparticles

The nanosilica sample was generated by a sol-gel technique using a modified process based on a previous study [25]. Sand washed and dried at ambient conditions for 48 hours. Subsequently, the material underwent the milling process in a planetary ball mill and was subsequently subjected to sieving, resulting in particle sizes ranging from 27 to 38 micrometers. The experimental procedure involved a blending of 20 grams of sand with 62.5 grams of sodium hydroxide and subjecting the mixture to 500ºC for 30 minutes. Then, the mixture was agitated by adding 250 milliliters of distilled water. Upon the occurrence of the reaction, a uniform solution formed. Subsequently, a steady addition of 6 M malonic acid is performed until the pH of the solution attains a value of 3, leading to the formation of a white gel. The silica gel was washed several times with distilled water until pH reaches a value of 7, followed by separation using filter paper and a vacuum pump, and dried in an electric furnace for a duration of 12 hours at 110 ºC. The solid samples underwent a process of crushing, resulting in the formation of a fine powder, which identified as nanosilica white powder. Finally, different characterization methods were used to characterize the obtained sample.

2.3. Ultrasonic treatment procedure

250 ml of crude oil sample was put in a container with a diameter of 7 cm and 9.7 cm in length. The ultrasonic probe was positioned at the center of the container, at a depth approximately equal to half of the oil sample's total volume. A rubber stopper was used to prevent the loss of the lighter molecules. A water bath was utilized to regulate the temperature during the experiments. Then, the sample was exposed to ultrasound waves at different ultrasonic powers (120, 240, 360, 480, and 600 watts) for various radiation times (1, 2, 3, and 4 minutes) at 0.6 duty cycle and 25 ºC. Then the sample at the best time and power was exposed to different duty cycles (0.2, 0.4, 0.6, and 0.8) and temperatures (25, 35, 45, and 55 ºC) to examine the influence of these variables on the viscosity

of crude oil. After that different concentrations of silica nanoparticles (500, 1000, 1500, and 2000 mg/L) were added to the crude oil at the best conditions. The temperature of the sample was determined by employing a thermocouple. Subsequently, the sample remained within a sealed container to reach equilibrium with the surrounding environment, facilitating the condensation of all volatile substances and the subsequent formation of a liquid phase. The experimental configuration is depicted in Fig. 1. The measurement of the viscosity of crude oil was conducted with a Brookfield DV-II+ Pro viscometer, according to the ASTM D2196 test method. Finally, to verify the findings, asphaltene content and sulfur content were performed on the original sample and the final samples under the best conditions, using an asphaltene analyzer (APD-600A) according to the IP-143 test method and a SINDIE OTG Sulfur Analyzer according to the ASTM D7039 test method to measure asphaltene and sulfur content, respectively.

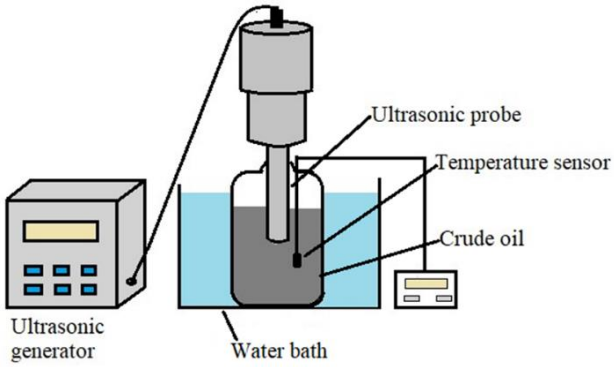


Fig. 1. The experimental setup

3- Results and discussion

3.1. Silica nanoparticles characterization

a. X-Ray diffraction analysis (XRD)

The X-ray diffraction pattern of NS reveals a large peak at 22.55º, which is a characteristic feature of amorphous silica products, as depicted in Fig. 2. The presence of a peak at 22.55º suggests the presence of silica, which is further supported by the findings of An et al. [26] and Nguyen et al. [27]. The absence of any other peaks is indicative of the quality of the product and there are no other crystalline impurities in the extracted silica.

b. Atomic force microscope (AFM)

The Atomic Force Microscope was used to investigate the morphology of prepared silica. The outcome of the scanned region reveals the topographical characteristics of silica particles, exhibiting an aspherical morphology and the presence of small particles in agglomerated arrangements. The presented data in Fig. 3 illustrates the range of particle sizes and the corresponding scanned area of silica nanoparticles. It indicates the prepared silica consisted of particles with diameters ranging between 10 and 110 nm, and the average diameter was 59.93 nm. These results agree with Jalil and Hussein [28].

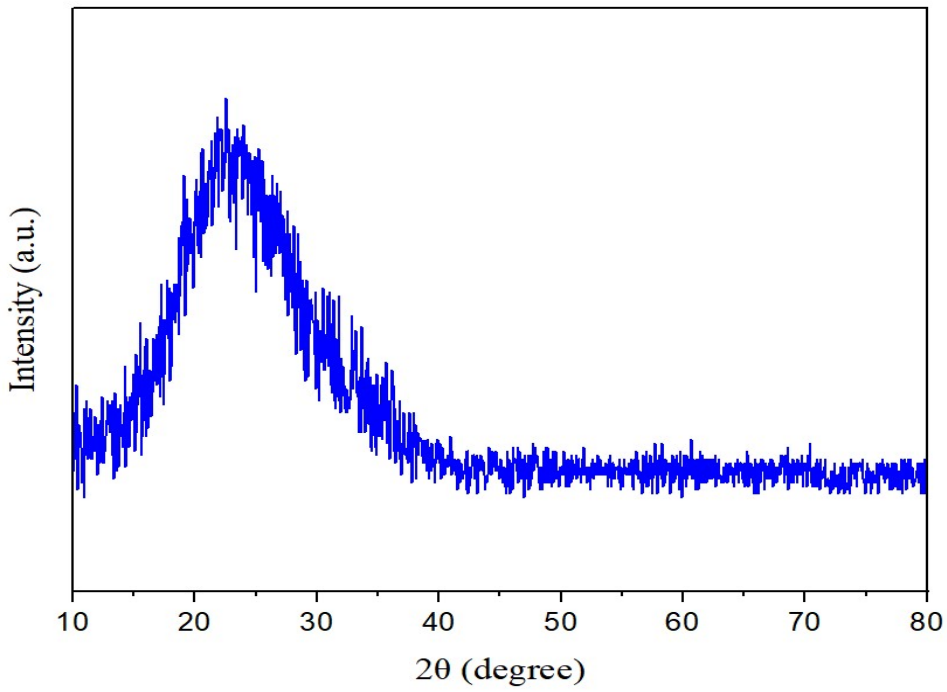


Fig. 2. XRD pattern of silica nanoparticles

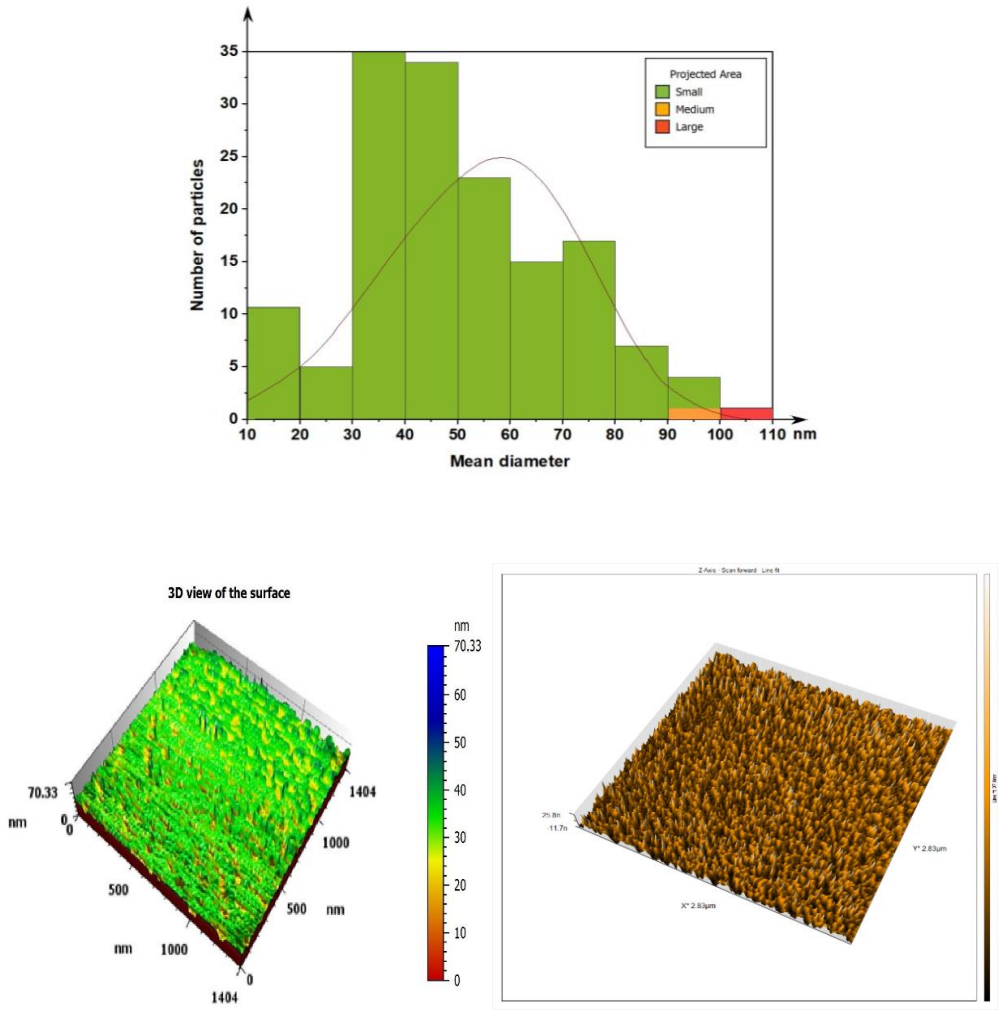
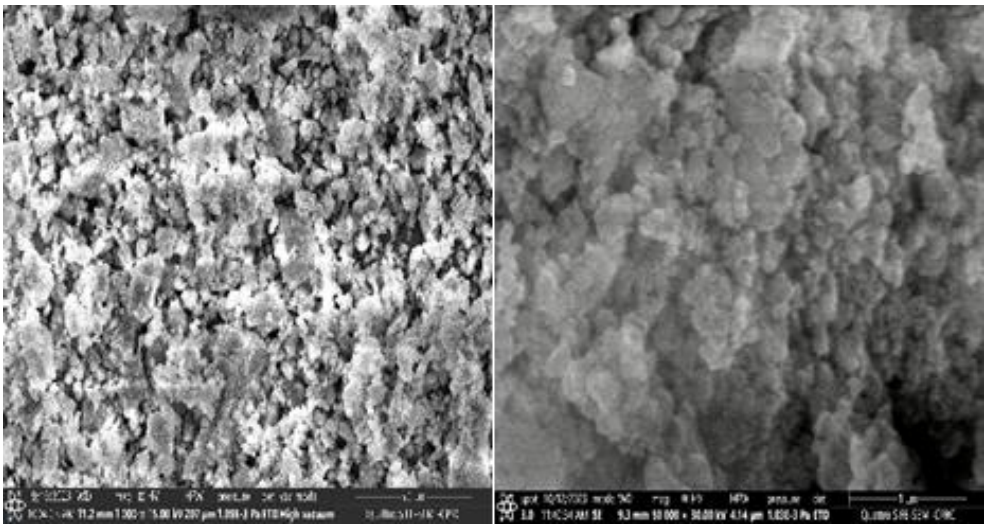


Fig. 3. AFM of nanosilica

c. Scanning electron microscopy (SEM)

SEM was employed to determine the morphology and distribution of the NS. Fig. 4 presents SEM pictures depicting the silica nanoparticles at various levels of optical magnifications of 50 µm and 1 µm. The micrographs clearly indicate an amorphous nature, and the particles show a notable tendency to aggregate, have a spherical morphology, and seem porous. This result is in agreement with Sharafudeen et al. [29].



(a) (b)

Fig. 4. SEM for prepared nanosilica by two magnifications (50 µm) in (a) and (1 µm) in (b)

d. BET surface area and pore volume

The surface area and pore volume of the synthesized silica nanoparticles were determined. The study reveals that nano silica possesses a surface area of 563.23 m2/g and a pore volume of 0.3522 cm3/g. Silica having these specifications exhibits suitability as a compound for utilization as a catalytic support or as an adsorbent. The surface area and pore volume determined in this study are greater than that found by Saleh et al. [30] and Rajan et al. [31].

e. Fourier transform infrared spectroscopy (FTIR) analysis

The FTIR spectra of the nano-silica (NS) are depicted in Fig. 5. The SiO2 spectra exhibit distinct peaks at 465.49 cm-1, which can be attributed to the bending modes of the siloxane (Si-O-Si) group. Additionally, the spectra display peaks at 796.18 cm-1 and 1090.30 cm-1, corresponding to the symmetric and asymmetric stretching vibrations of the siloxane group, respectively. The presence of -OH groups within the sample is indicated by a broad absorption band observed at 3441.48 cm-1 and a peak at 1634.29 cm-1. The presence of a significant number of silanol groups (Si-OH) is observed at a wavenumber of 954.12 cm-1. The findings presented are consistent with Okoronkwo et al. [32], Foroutan et al. [33].

f. Thermogravimetric analysis (TGA)

The performance of nanosilica particles was evaluated by subjecting them to a temperature range of 25-800 ◦C. The variation in weight loss of the sample was recorded as a dependent variable with respect to temperature, as depicted in Fig. 6. The observed reduction in weight for NS is around 8.61% within a range of 25 to 200 ◦C, which could potentially be ascribed to the elimination of water molecules present inside the pores of NS. During the second stage, a weight loss of around 7.01 is seen within a range of 200 to 755 ◦C. This weight loss can be attributed to the condensation of silanol groups, leading to the elimination of water molecules that were previously produced. Hence, it was observed that the silica nanoparticles required an extended duration of heating in order to release water molecules from the nanosilica sample. The decrease in mass losses of the sample exhibits a gradual trend as temperature increases. No further decrease in weight was seen after reaching a temperature of 755 ◦C. This finding confirms that nano silica powder exhibits good thermal stability.

3.2. The analysis of viscosity reduction

a. Effects of ultrasonic power and irradiation time

Fig. 7 (a-d) shows the influence of ultrasonic power of 120, 240, 360, 480, and 600 W for different exposure times of 1, 2, 3, and 4 min at 0.6 duty cycle and 25 ºC on the percentage of viscosity reduction of heavy crude oil.

The higher percentage of reduction in viscosity of crude oil at 1 min of exposure time (Fig. 7 a) was 9.25% at 480 W due to the production of ultrasonic energy, which may lead to the breaking of intermolecular connections and the separation of hydrocarbons from other particles, which may account for a decrease in the viscosity of crude oil [33,34], but at 600 W the reduction in viscosity decreased due to the rise in temperature of the crude oil caused by the delivery of energy to the system [35], resulting in the evaporation of the lighter components.

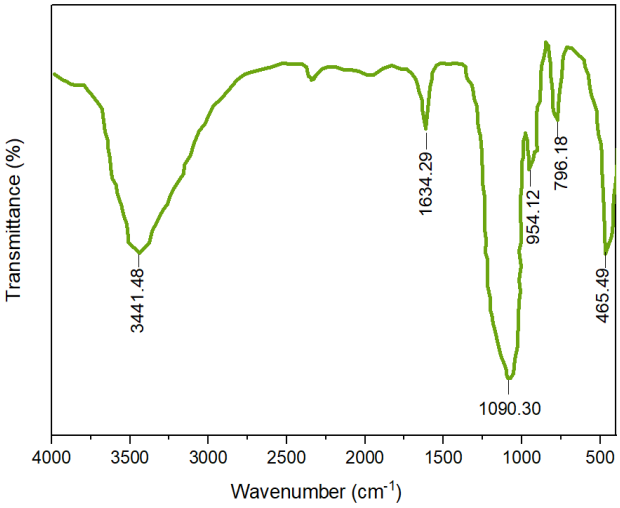


Fig. 5. FTIR spectra of prepared NS

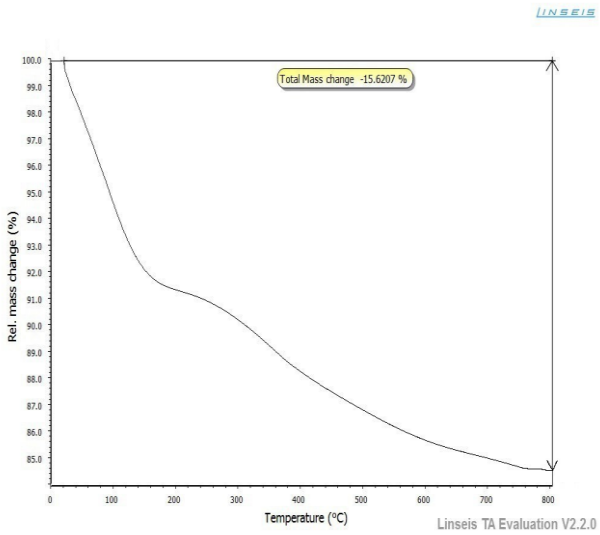


Fig. 6. TGA of prepared silica nanoparticles

From Fig. 7 b, which shows the impact of ultrasonic power on the reduction in viscosity at 2 min of irradiation, the maximum degree of viscosity reduction was 14.29% at 360 W, but when the power increased, the viscosity increased more. It can be noted that one minute of irradiation was not enough, so high energy was required to reach the maximum reduction, when the time raised to 2 min, the reduction in viscosity increased at an ultrasonic power lower than that of 1 min.

At 3 min of ultrasonic irradiation (Fig. 7 c), the maximum reduction in viscosity was 9.08% at 360 W. By

comparing this result with that from Fig. 7 (b), it could be noticed that when the time was raised to 3 min at 360 W, the rate of reduction in viscosity decreased as a result of asphaltene breaking into smaller broken particles. Moreover, there is evidence of the synthesis of heavy molecules with additional branches [36].

Fig. 7 d shows a 6.39% reduction in crude oil viscosity at 4 min and 240 W. This indicates that the required power to reach the maximum reduction decreased as time increased. By comparing the results, a higher reduction in viscosity (14.29%) was obtained at 2 min and power of 360 W, which was the best conditions, but after these conditions, the viscosity exhibited a higher value compared to its initial viscosity for some samples.

b. Effect of duty cycle

The impact of the duty cycle on the viscosity reduction of crude oil was studied by exposing crude oil samples to ultrasonic waves at 0.2, 0.4, 0.6, and 0.8 duty cycles at 2 min, 360 W, and 25 ºC, as shown in Fig. 8 which indicates that when the duty cycle increased, the degree of viscosity reduction increased, which reached 15.4% at the 0.8 duty cycle. This decrease in viscosity is a result of the enhanced impact of the ultrasonic wave and the subsequent formation of bubbles, which is facilitated by increasing the duty cycle [37].

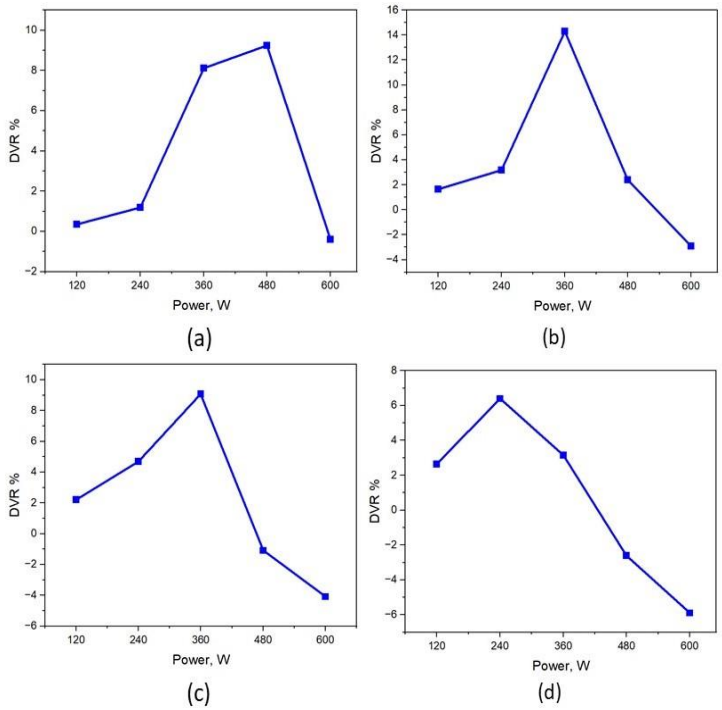


Fig. 7. The influence of (a) 1 min, (b) 2 min, (c) 3 min, and (d) 4 min of irradiation time at different ultrasonic power, 0.6 duty cycle, and 25ºC on the degree of viscosity reduction percentage of heavy crude oil

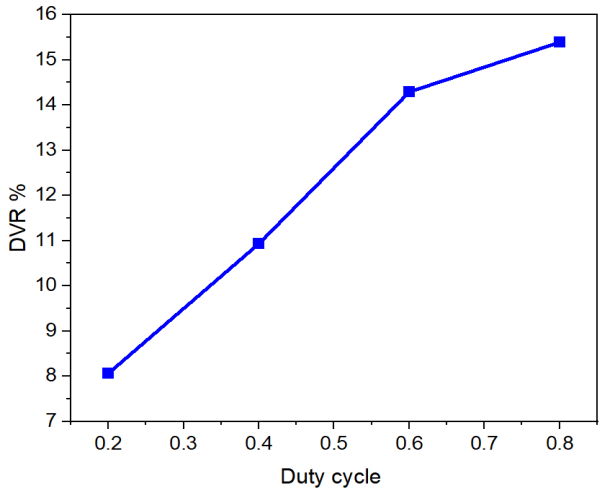


Fig. 8. The influence of duty cycle on the degree of viscosity reduction percentage of heavy crude oil at 2 min of ultrasonic irradiation, a power of 360 W, and 25°C

c. Effect of temperature

The impact of temperature on the reduction in viscosity of crude oil was studied by exposing crude oil samples to ultrasonic waves at temperatures of 25, 35, 45, and 55 ºC, with 2 minutes of irradiation, power output of 360 W, and a duty cycle of 0.8, as shown in Fig. 9. The data presented in this figure demonstrates that when the temperature rose to 35 ºC, there was a corresponding increase in the degree of viscosity reduction, reaching a value of 20.81%.

However, it was noted that when the temperature increased more than 35ºC, the extent of viscosity reduction exhibited a drop. This behavior can be clarified by the thermal-induced boiling effect and the occurrence of cavitation, which promotes the vaporization of less dense components [10].

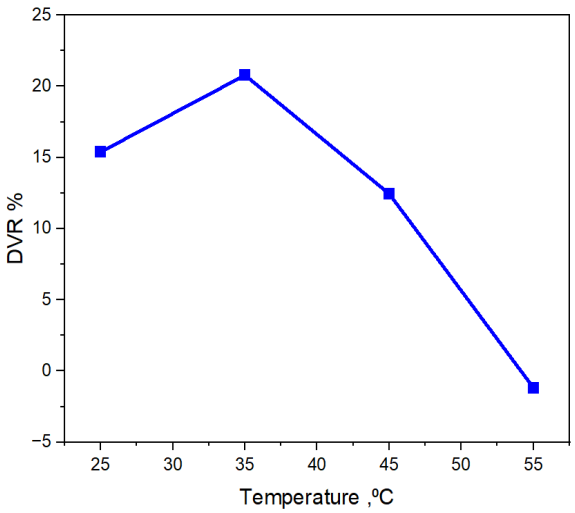


Fig. 9. The influence of different temperatures on the degree of viscosity reduction percentage of heavy crude oil at 2 min of ultrasonic irradiation, a power of 360 W, and 0.8 duty cycle

d. Effect of silica nanoparticles concentration

The crude oil samples were irradiated at 2 min, 360 W, 0.8 duty cycle, and 35 ºC with the addition of different concentrations of NS to examine their influence on the viscosity of crude oil. It can be noted in Fig. 10 that as the nanoparticles concentration increases, there is a drop in viscosity. This continues until it reaches the best concentration (1500 mg/L). At this concentration, the reduction in viscosity was 27.83%. The observed results can be related to the phenomenon of asphaltenes adsorption, which leads to the disruption of the viscoelastic network of asphaltenes and consequently enhances the flow capacity of crude oil .

After the best concentration, there was an observed rise in the viscosity of the oil. A rise in nanoparticle concentration resulted in an increase in the molecular bonds between the nanoparticles and the adsorbed asphaltene, facilitating the aggregation of larger particles and a subsequent rise in the viscosity of the oil [36]. These results demonstrate that the use of nanoparticles can effectively augment the reduction in viscosity of heavy oils by ultrasound waves.

3.3. Asphaltene and sulfur content reduction

To validate the results of the experiments, in addition to the viscosity an analysis of asphaltene content, and sulfur content was conducted on both the initial sample and the final samples under the best conditions. The purpose of these analyses was to examine the phenomenon of lightning and cracking in the bonds of hydrocarbons found in crude oil .

According to the data presented in Fig. 11, the asphaltene concentration exhibited a decrease from 6.379 wt% to 5.227 wt% after the application of ultrasonic treatment. This decline can be attributed to the structural degradation of the asphaltenes, leading to their breakdown into lighter constituents. This phenomenon is closely associated with the cracking of the alkyl side chain. The asphaltene content decreased to 4.482 wt% after the introduction of 1500 mg/L of NS. This finding provides evidence that Silica nanoparticles can adsorb asphaltenes. Furthermore, it is possible to reduce the formation of asphaltene particles and positively impact the rheological characteristics of the oil [38].

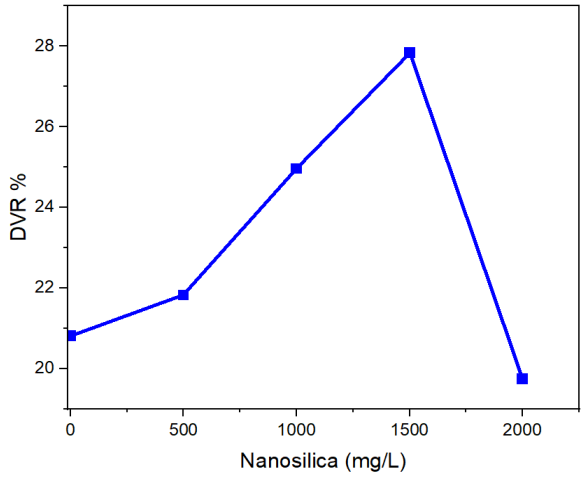


Fig. 10. The influence of different concentrations of NS on the degree of viscosity reduction percentage of heavy crude oil at 2 min of ultrasonic irradiation, a power of 360 W, 0.8 duty cycle, and 35 ºC

As shown in Fig. 12, the sulfur content reduced from 4.68 wt% to 4.30 wt% after the ultrasonic treatment because the cavitation process leads to the breaking of C-S, C-C, C-O, and C-N bonds in heavy oil, as a result of the significant rise in local temperature caused by the collapsing bubbles .

According to [39], adding a catalyst to the reaction system can reduce the bond energy, which makes it easier for ultrasonic waves to break the C-S bond. Therefore, a decrease to 4.12 wt% was observed in the sulfur content after adding 1500 mg/l of NS.

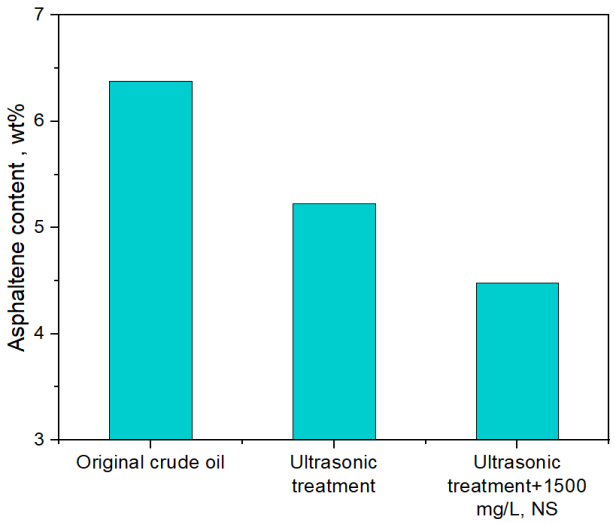


Fig. 11. The asphaltene content in heavy crude oil before and after the ultrasonic irradiation process with and without NS addition

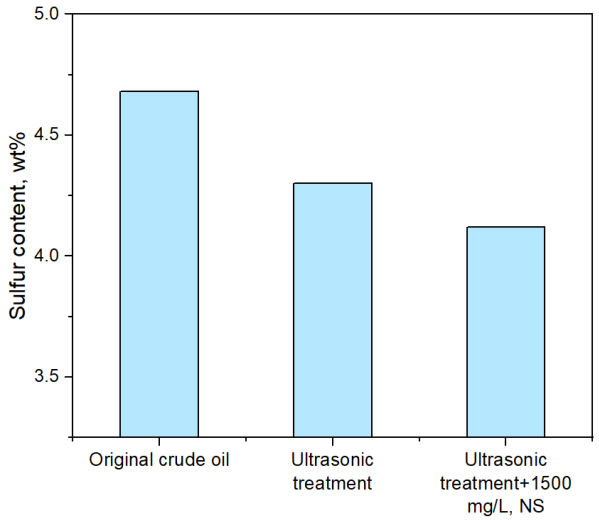


Fig. 12. The sulfur content in heavy crude oil before and after the ultrasonic irradiation process with and without NS addition

4- Conclusion

The preparation of high-purity silica nanoparticles from sand using the sol-gel method is both environmentally favorable and cost-effective because of the availability of high-grade sand in western Iraq. Furthermore, the highest surface area (563.23 m^2/g) was obtained using an organic acid, specifically malonic acid. The combined use of both ultrasonic technology and NS resulted in a more efficient reduction in crude oil viscosity than using ultrasonication alone. Under moderate conditions of an ultrasonic power of 360 W, a two-minute treatment duration, a duty cycle of 0.8, and a temperature of 35 °C, the viscosity of the crude oil sample decreased by 20.81%. This reduction further increased to 27.83% when 1500 mg/L of prepared NS was added, due to its ability to adsorb the asphaltene particles. The findings indicated that as the time increased more than the best time, the viscosity reduction rate decreased, this phenomenon could be attributed to the re-association of heavy oil molecules. Also, an increase in temperature and ultrasonic power resulted in an elevation of the viscosity of the crude oil, which can be related to the phenomenon of boiling.

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دراسة تأثير الموجات فوق الصوتية والسليكا النانوية في تخفيض لزوجة خام شرق بغداد الثقيل

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الخلاصة

يدرس البحث الحالي التأثيرات المشتركة للموجات فوق الصوتية والسليكا النانوية (NS) على تقليل لزوجة النفط الخام الثقيل الذي له قيمة API تبلغ ٢٠,٣٢. تم تصنيع جسيمات السليكا النانوية بمتوسط حجم ٥٩,٩٣ نانومتر ومساحة سطحية ٥٦٣,٢٣ م٢/جم باستخدام تقنية Sol–Gel من الرمال العراقية. يشير تحليل XRD إلى وجود سليكا غير متبلورة. أظهر SEM أن NS يُظهر ميلاً للتكتل. يُظهر أطياف FTIR وجود مجموعات siloxane و silanol. بالإضافة إلى ذلك، أظهر تحليل TGA فقداناً إجمالياً للوزن بنسبة ١٥,٦٢%، مما يثبت الاستقرار الحراري لـ NS. تضمنت التجارب دراسة تأثير طاقة الموجات فوق الصوتية وزمن التعرض ودورة التشغيل ودرجة الحرارة والتأثيرات المشتركة للموجات فوق الصوتية وجسيمات السليكا النانوية على درجة تخفيض لزوجة النفط الخام الثقيل. أظهرت النتائج أن لزوجة النفط الخام الثقيل انخفضت بنسبة ٢٧,٨٣% عند دقيقتين من التعرض للموجات فوق الصوتية، طاقة ٣٦٠ واط، ٠,٨ دورة عمل، ٣٥ درجة مئوية، بمساعدة السليكا النانوية بتركيز ١٥٠٠ ملجم/لتر.

الكلمات الدالة: الأسفلتين، النفط الخام الثقيل، تقليل اللزوجة، الموجات فوق الصوتية، النانوسليكا.