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Removal of Wax Deposit from Tight Formation Using Ultrasonic Cavitation with Thermochemical Heat Source

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Abstract

Formation damage phenomenon constitutes serious operational and economic problems to the petroleum production. Oil production in certain reservoirs is inadvertently impaired by precipitation and deposition of the high molecular weight components such as paraffin wax. A facile applicability of synergistic effects of thermochemical reaction and ultrasonication to mitigate wax deposition has been presented in this article. Thermochemical heat source has to do with exothermic heat generation from certain chemical reactions. On the other hand, ultrasonication causes cavitation and implosion of bubbles, which is trasimmted as energy in the medium and assit in detaching contaminants from the surface. Series of imbibition experiments were conducted at different ultrasound frequencies (low 28kHz, and high 40kHz), exposure times (20, 40, and 60 mins), and different molarities (M1, M2, and M3) of the thermochemical fluids (TCF), for removing wax deposit from tight Scioto Sandstone core samples. The performance was followed through permeability and porosity tests, as well as Scanning Electron Microscopy with Energy-Dispersive X-ray (SEM-EDX) analyses. Ultimately, the results revealed promising potentials for the proposed technology for efficient paraffin wax removal from a tight rock sample up to 70% within the experimental limits investigated.

Keywords: Formation damage, Wax deposition, Thermochemical, Ultrasonication, Optimization

Introduction

Overview

Under certain reservoir conditions, the heavy component of crude oil such as paraffin wax could crystallize, plug the pore space, and ultimately reduces formation permeability (Venkitaraman et al., 1995; Wong et al., 2003). The search for effective methods to mitigate formation damage has become imperative due to operational and economic concerns that these can cause (Alade et al., 2020). Wax deposition problems are most efficiently addressed using thermal, mechanical, and chemical methods (Persad, 2018). The chemical treatment method, although effective (Ragunathan et al., 2020), is usually affected by several challenges including health and safety implications, environmental concerns, and cost, since large quantity of solvent is usually required. Typically, the chlorinated hydrocarbons are often efficient for treatment of wax deposition in production strings and may also be applied to remediate formation damage (Fan et al., 1996). However, a major concern is the treatment and disposal of the produced fluid. The thermal method, including steam, hot water, or hot oil injection is an efficient method for wax removal due to wax dissolution at higher temperature. However, hot fluids can become less effective due to heat loss (Alade et al., 2020).

The objective of this research is to investigate the efficiency of a new method of wax damage remediation using synergistic effect of ultrasonic cavitation and heat generated from reaction of thermochemical fluids (TCF). The TCF would be injected and activated at the formation temperature, so that the heat loss can be miminzed. In addition, since the heat generation is coming from the reaction itself, the cost of energy as well as emission of the greenhouse gases (GHG) would be significantly reduced. On the other hand, the power ultrasound would enhance the process by improving heat and mass transfer and enhancing the reactivity of the TCF. The proposed treatment can also find useful applications in treating other organic scale deposition such as aphaltene.

Thermochemical heat source

Thermochemical reactions are those that involve exothermic release of heat to the surroundings due to higher energy potential (i.e., $\Delta H < 0$: see Figure 1). Compared to the conventional steam generation process, the thermochemical process of steam generation offers the advantage for lower capital and/or operating costs, higher overall thermal efficiency, and abatment of environmental pollution due to emission of GHG such as CO_2 (Alade et al., 2019a). Thermochemical heat source had been recently introduced as an advancement in thermal stimulation techonology, owing to significant heat and pressure, which could be generated from the exothermic reactions of certain chemical reactants (Al-Nakhli et al., 2016; Alade et al., 2019b). The thermochemical stimulation process has been used in treating several formation damage problems including wax deposition (Leiroz et al., 2005; Mahmoud, 2019). Sufficient temperature increase (up to 260 °C) and pressure (up to 2000 psi) could be accessed from a typical thermochemical reaction involving ammonium chloride and sodium nitrite (Al-Nakhli et al., 2016). As shown in Equation 1, the process is friendly to handle since brine (NaCl) as well as Nitrogen gas is generated alongside steam from the reaction.

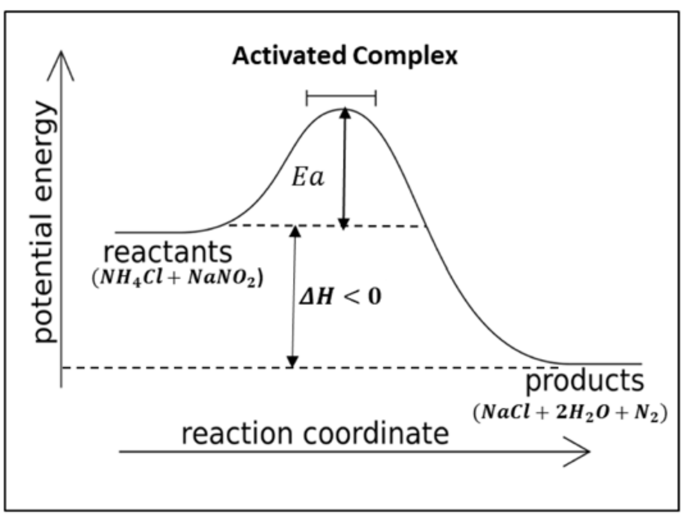


Figure 1—Energy level diagram of a typical exothermic reaction (E_a is the activation energy and ΔH is the enthalpy of reaction: Alade et al. (2020))

$$NH_4Cl + NaNO_2 \rightarrow NaCl + 2H_2O + N_2 + \Delta H(heat) \tag{1}$$

The enthalpy changes of the system, ΔH, has been calculated as 370 $KJmol^{-1}$, with the reaction order $n = 1$ and the activation energy, E_a, ≈ 35.5 $KJmol^{-1}$ (Alade et al., 2019b). Moreover, from previous investigations (Hassan et al., 2018), it has been observed that the TCF injection is free from rock/fluid compatibility issues and the associated negative effects of precipitates compared with some other stimulation fluids. In addition, using the NMR pore analysis, it was revealed that TCF injection instigated pressure pulses that resulted in tiny fractures created in the core samples after flooding operations. Furthermore, thermochemical treatment has be found to increase the permeability of the treated core sample from 120 – 800 mD, while the porosity increased from 20 – 21.5%, after treatment.

Ultrasonic removal of contaminants

Exposure of particles and particulate structures to ultrasounds finds relevant applications in various fields (Knoop and Fritsching, 2014; Aktij et al., 2020). The interactions of high-intensity sound fields with homogeneous or heterogeneous media involves either physical mass transport or chemical production of radicals, excited species, and single electron transfer. This induces interesting phenomena including acoustic cavitation and acoustic streaming (Knoop and Fritsching, 2014). The acoustic cavitation process leads to expansion and violent implotion of gas or vapor-filled cavities in fluids, which produces localized spots of high temperature and pressure. The violent collapse of the oscillating bubbles during acoustic cavitaion has important applications in removing particulate contamination from hard surfaces (Venkitaraman et al., 1995). The acoustic streaming, on the other hand, enhances fluid agitation and is effective for liberating particles attached to surfaces. The simplified mechanism of ultrasonication showing microbubble generation and implosion is shown in Figure 2.

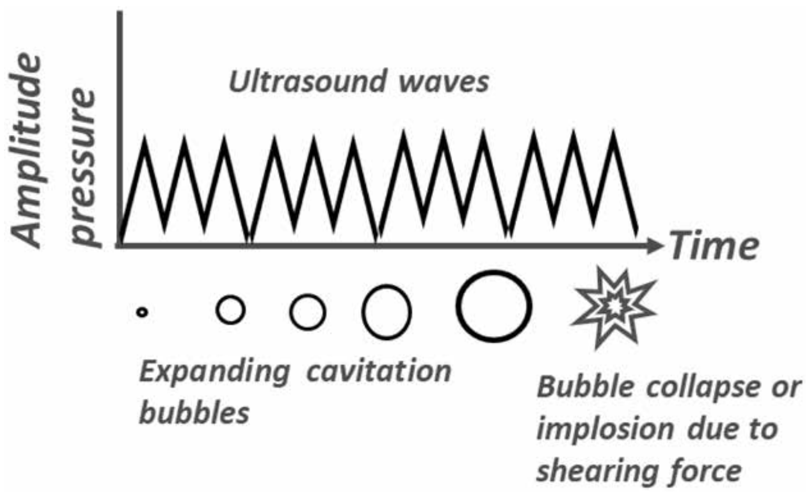


Figure 2—Illustration of ultrasonication mechanism

Impressive efforts have been made towards developing ultrasonic technology for various applications in petroleum industry such as enhanced oil recovery, oil sand extraction, demulsification, viscosity reduction, oily wastewater treatment and oily sludge treatment (Vakilinia, 2012; Hamidi et al., 2012; Mullakaev et al., 2015; Li et al., 2020; Luo et al., 2021). Although an old concept, the power ultrasound technology represents an emerging grenner technology in formation damage mitigation compared with the conventional well stimulation methods (Wong et al., 2003; Mullakaev et al., 2015; Li et al., 2020). The acoustic well stimulation offers several advantages including opportunity for underbalanced stimulation, real time monitoring and optimization, improved removal of fines damage; removal of drilling mud filter cakes; cleaning of gravel packs, screens, slotted liner, perforations and near wellbore formation, eliminating the use of complex chemicals and fluids, wireline (or CT) conveyed treatment, precise zonal control, and enhanced chemical treatments (Wong et al., 2003). Promising results have been reported from different applications of high frequency ultrasonic wave technology including asphaltenes flocculation inhibitor (Amani et al., 2015), removal of inorganic scale depositions including NaCl and KCl salts from the near-wellbore regions (Jaber et al., 2017), removal of unwanted submicrometersized solids from drilling fluids (Wang et al., 2021), matrix acidizing (Khaldi et al., 2021), removal of near-wellbore paraffin wax deposits and polymers (Roberts et al., 2000), mud cake removal and mud filtration treatment (Venkitaraman et al., 1995; Vakilinia, 2012).

Methodology

The methodology employed includes preparation of damaged core samples by saturation with paraffin wax, imbibition experiments for cleaning rock samples using thermochemical fluids under ultrasonic condition, and evaluation of the efficiency using rock petrophysical tests and SEM-EDX analyses. Figure 3 is the schematic illustration of the imbibition experiment.

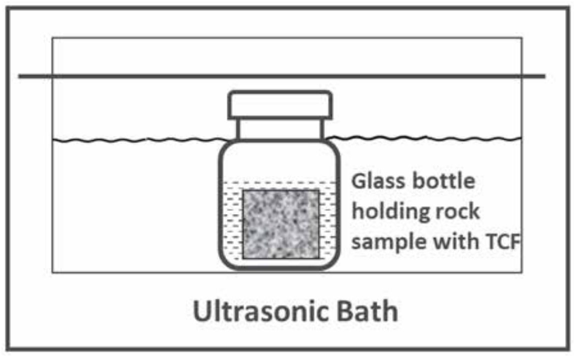


Figure 3—Schematic illustration of the imbibition experimental arrangement in an ultrasonic bath.

Preparation of rock samples

Scioto tight Mississipian Sandstone rock samples with a permeability of ≈0.01 mD and a porosity ranging between ≈17% were used in these experiments. The permeability of the core samples was determined using a steady-state bench top permeability system (BPS 350, Porous Materials), while the porosity was determined using the helium porosimeter based on Boils Law (He Porosimeter HEP-P, Vinci Technologies). The field sample of paraffin wax was supplied by an Arabian Oil Company. Before wax saturation, the rock samples were held in a transfer cell and vacuumed for sufficient time to remove the air contaminant. The wax sample was heated to 60 °C to allow liquefaction for ease of saturation. The saturation was done at the same temperature at the injection rate of 0.5 cc/min. Then, the core samples were held at 2000 Psi for 72 hours to ensure complete saturation. Subsequently, the samples were left inside the coreholder for one week before the experiments started.

Wax cleaning experiments

The wax cleaning experiments were conducted using the Branson ultrasonic bath (Model 8800). The equipment was operated at low and high frequency modes with 28 and 40 kHz, respectively. The stock solution of thermochemical fluids (TCF) was supplied by an Arabian Oil Company. The physical properties including temperature and pressure changes obtained from 20 ml total reacting volume of the samples are given in Table 1.

Table 1—Properties of the TCF Samples

Sample/Molarity	Surface Tension (mN/m)	Density (g/cc)	μ (cp)	ΔT (°C)	ΔP (Psi)
TCF_1	37.17	1.0503	1.0293	38	230
TCF_2	37.18	1.0371	1.1533	45	270
TCF_3	32.88	1.1465	1.5317	60	305

Series of imbibition experiments were conducted at room temperature (21 °C) under the low and high frequencies by soaking the damaged rock samples in different TCF solutions (Molarity 1 – 3). The exposure time was operated between 20 – 60 mins. The optimum conditions for operation were obtained through empirical modeling and response surface methodology using the Central-Composite Experimental design. The experimental information is detailed in Table 2. The neutral runs were those conducted on the bench-top without exposure to ultrasonication.

Table 2—Input Vraiables used in Central Composite Design

Factor	Name	Units	Minimum	Maximum	Coded Low	Coded High	Mean	Std. Dev.
A	Molarity	M	1.0000	3.00	-1 ↔ 1.00	+1 ↔ 3.00	2.00	0.6882
B	Time	min	20.00	60.00	-1 ↔ 20.00	+1 ↔ 60.00	40.00	13.76
C	Frequency	KHz	Neutral	High			**Levels:**	3.00

Evaluation of cleaning efficiency

The performance of the system was evaluated through mass balance by comparing the weight of the treated samples, with the weight before and after wax damage. In addition, the efficiency of the optimum treatment conditions was evaluated using the Scanning Electron Microscopy (JCM-700 NeoScope Benchtop SEM) equipped with energy dispersive X-Ray spectroscopy (SEM-EDX) to determine the elemental distribution in the selected rock sample before and after treatments.

Results and Discussion

During ultrasonication process (Venkitaraman et al., 1995), the detachment of particles, which is stuck at the pore walls and throats will occur when the drag force created from the passage of a wave overcomes the attractive force holding the particle in place. Ultimately, the process would improve the effective permeability. Moreover, thermochemical reaction essentially removes wax deposit through temperature increase (Alade et al., 2020). At typical reservoir conditions, the temperature change, presented in Table 1, is sufficient to dissolve wax crystal since it is above the pour point. More so, the pressure change, and the surface activity of the chemical are good indications for the ability of the chemical to alter the rock wettability and assist in wax removal. Figure 4 compares the percentage wax removal from the experiments. It generally shows that the wax removal increases with increasing molarity of the TCF as well as reaction time. This observation, as expected, has to do with the reactivity of the fluids, as explained above. Notably, the temperature change and pressure generated from the samples varried as 38 °C (230 Psi), 45 °C (270 Psi), and 60 °C (305 Psi) from M1, M2, and M3, respectively, as presented in Table 1. In addition, it was observed that wax removal was improved with exposure to ultrasound compared with the neutral experiments (runs without ultrasonication). This observation can be linked with the conversion of the reaction in the absence of ultrasound. Our previous findings (Alade et al., 2019b) revealed that the conversion of the reaction is normally withing 10 minutes. However, from the preliminary unreported observations, it has been observed that the reaction can progress significantly beyond this period, when exposed to the ultrasound power. Theoretically, these results corroborate the excitation of particles and formation and growth of microbubbles with higher collision energy, as expected. Furthermore, the removal of wax does not increase significantly between TCF molarities M2 and M3 at high ultrasonication frequency (*High = 40kHz*), and notably at longer exposure time (60 minutes), as compared with the neutral experiments. Although not much is clearly known about the meachnism of thermochemical reaction under ultrasonication at this moment, it could be suggested that the reactivity and/or bubble cavitation action might have been suppressed in reaction using M3 at high exposure frequency, since this reaction would generate higher pressure, as given in Table 1.

This theory probably is corroborated by the observation by Venkitaraman et al. (1995), that when the fluid pressures were higher than acoustic pressure, the bubble cavitations were observed to be suppressed.

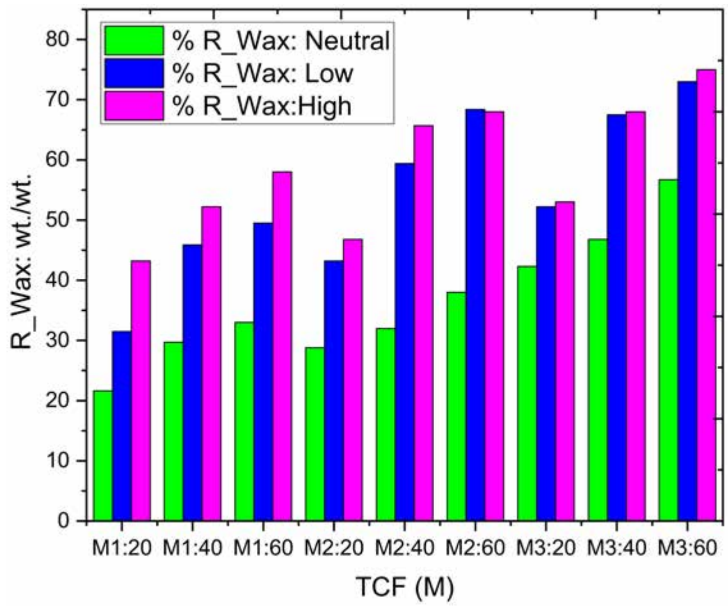


Figure 4—Percentage wax removed using TCF solutions at different ultrasonic frequencies (Low = 28kHz; High = 40kHz; and Neutral = Reference) and exposure times (20, 40, 60 minutes)

At this premilinary stage of the investigation, the focus to search for the optimal operating conditions has been informed by the desire to manage the reaction for improved performance. The experimental design approach essentially generates polynomial equations that can be used to predict the performance of the process under different conditions, subject to the limit of the experiments. As presented in Equations 4, 5, and 6, the model expresses the inverse of the dependent variable (% wax removal, R_wax) in terms of the experimental input variables viz. TCF molarity (M), and exposure time (T), under different ultrasound frequencies, i.e., neutral (no exposure), *low (28kHz),* and *high (40kHz)*). It also shows that the variables have some interactive effects between each other on the performance of the system.

Equation 4: Neutral Frequency Condition

$$\frac{1}{(R_{WAX})} = 0.072 - 0.009(M) - 9.5E - 4(T) + 1.31E - 4(M*T) - 0.0011(M^2) + 5.27E - 6(T^2) \quad (4)$$

Equation 5: Low Frequency Condition

$$\frac{1}{(R_{WAX})} = 0.058 - 0.014(M) - 8.26E - 4(T) + 8E - 5(M*T) + 1.6E - 3(M^2) + 5.52E - 06(T^2) \quad (5)$$

Equation 6: High Frequency Condition

$$\frac{1}{(R_{WAX})} = 0.037 - 0.0048(M) - 4.76E - 4(T) + 4.9E - 6(M*T) + 6.1E - 4(M^2) + 3.85E - 06(T^2) \quad (6)$$

Figure 5 is the correlation of the experimental (actual) and predicted values of the wax removal (inverse of %R_Wax). The graph shows the acceptability of the model to predict the system. The Predicted R^2 is 0.88 and in reasonable agreement with the adjusted R^2 of 0.94; i.e. the difference is less than 0.2. Moreover, as shown in Figure 6, a desirable optimal conditon exists at TCF molarity of 3 (M3), 45 minutes exposure time, under low frequency (28kHz), to yield %69.5 of wax removal.

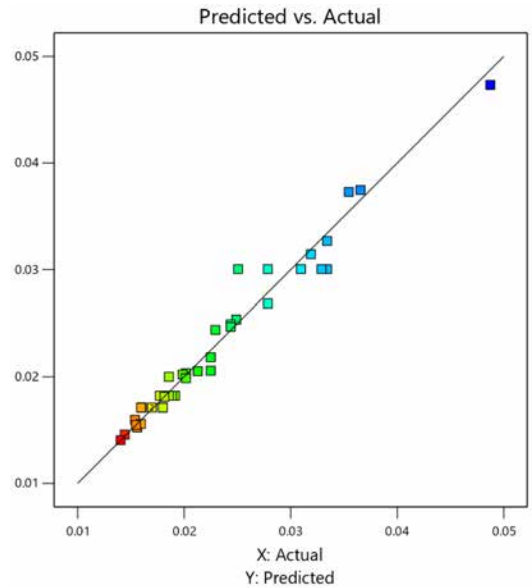


Figure 5—Predicted and experimental values of wax removal

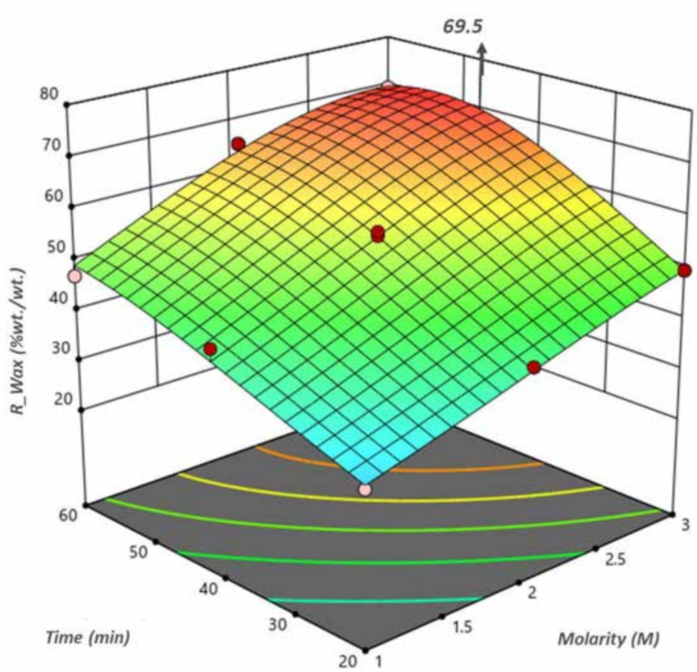
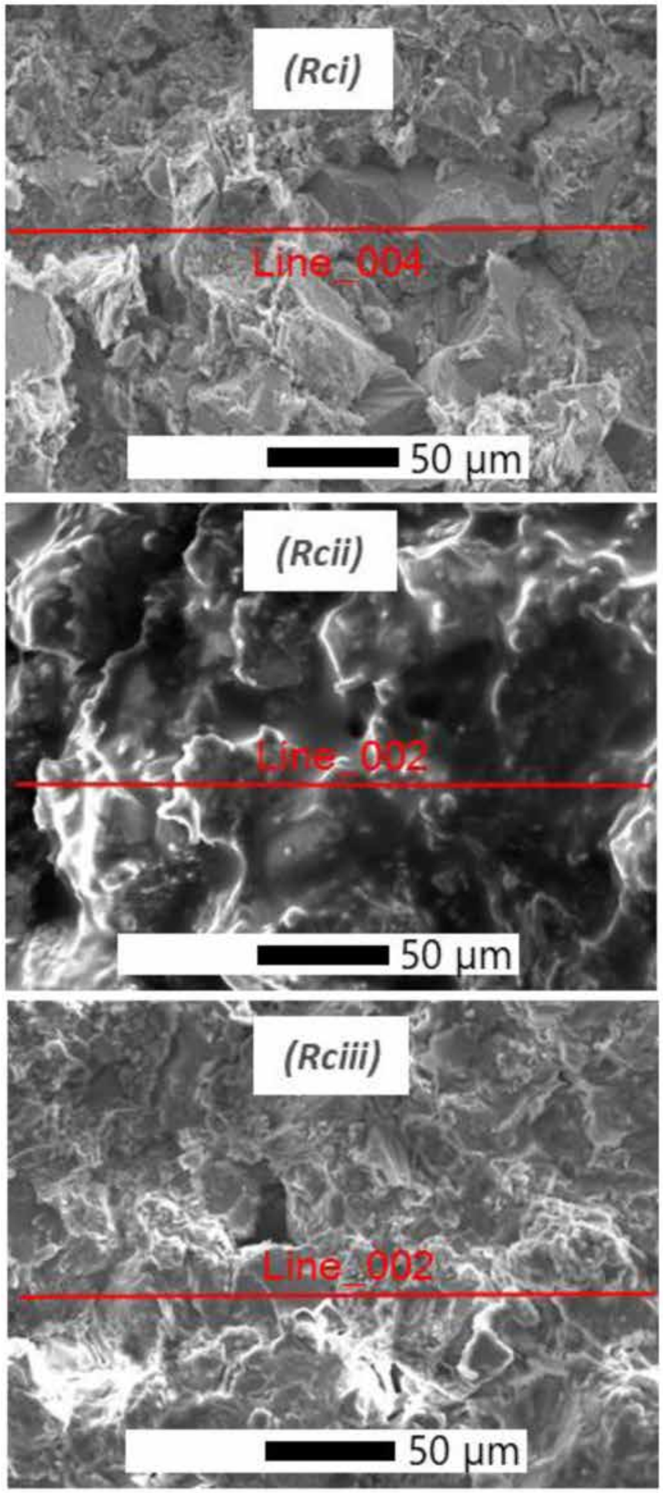
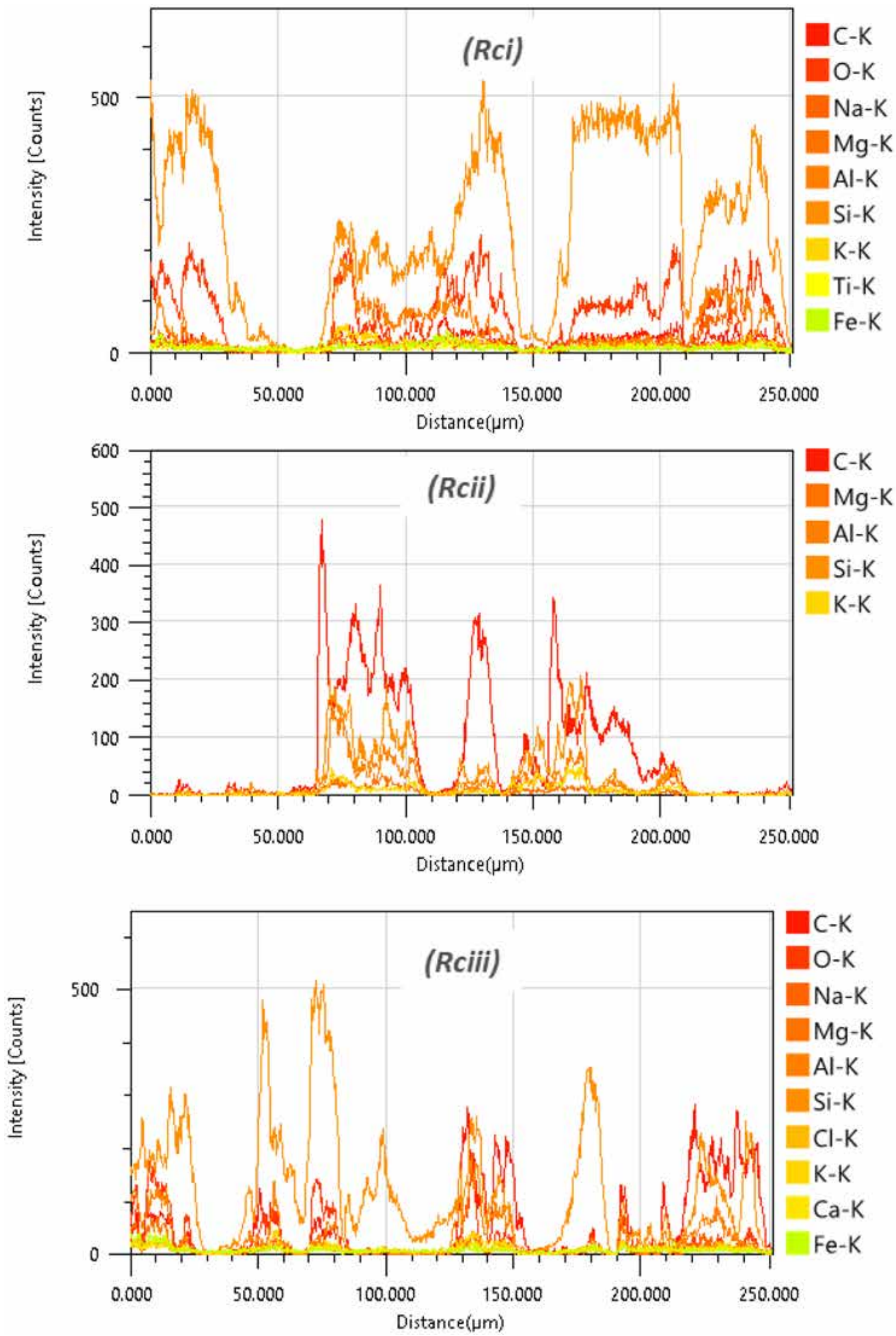


Figure 6—3D surface graph of wax removal under low frequency ultrasound (28kHz)

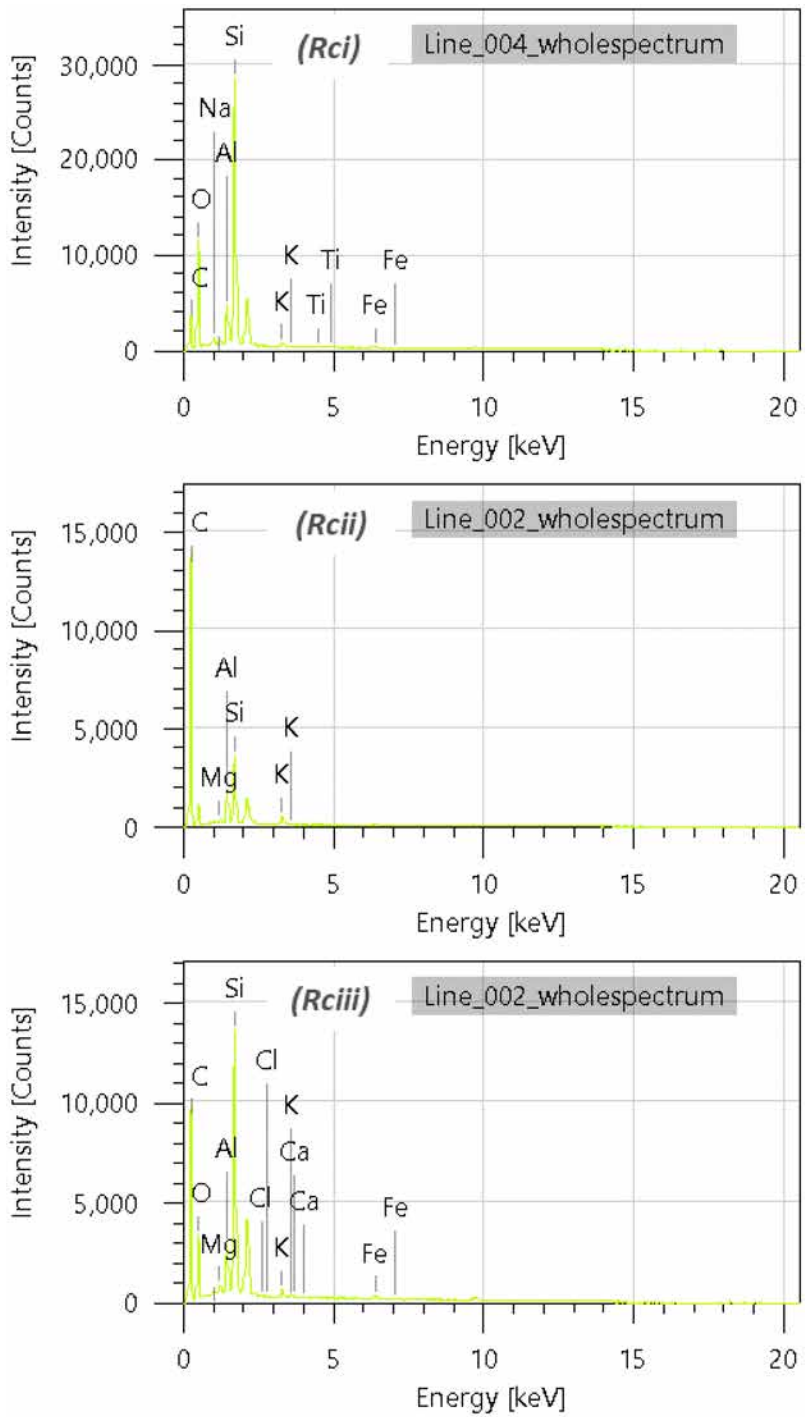
The SEM-EDX results are presented in Figures 7 – 9. Figure 7 presents the magnified SEM image of the original rock sample (*Rci*), the damaged rock sample (*Rcii*), and the treated rock sample (*Rciii*) using the optimum conditions. By comparing *Rci* and *Rcii,* a relatively opaque or blurring vision indicates formation damage due to wax deposition. Conversely, by comparing *Rci* and *Rciii,* a clearer image is an indication for significant wax removal fom the surface. More so, from Figure 8, the elemental distributions in *Rci* and *Rciii* are visually similar. In comparison, significantly less elements are visually available in *Rcii* compared with the former. Further, the elemental intensity shown in Figure 9 shows that a significant improvement to restore the damaged rock samples have been achieved after the treatment. From both of Figures 8 and 9, the availability of Chloride element in *Rciii* must have emanated from the TCF reaction, which produces brine (NaCl), as presented in Equation 1.



Figures 7—Scanning Electron Microsopy (SEM) image of original rock sample (Rci), the damaged rock sample (Rcii), and the treated rock sample (Rciii) using the optimum conditions.



Figures 8—Elemental distribution in the original rock sample (Rci), the damaged rock sample (Rcii), and the treated rock sample (Rciii) using the optimum conditions.



Figures 9—Elemental avaliability in the original rock sample (Rci), the damaged rock sample (Rcii), and the treated rock sample (Rciii) using the optimum conditions.

Moreover, the efficiency of the treatment method has been quantitively presented in Table 3 from the petrophysical parameters of the samples. The table shows that both the permeability and porosity of the original rock samples have been significantly impared due to wax deposition, as expected. From the *Rci* and *Rcii*, the permeability ratio after wax damage is 0.01, which is a significant indication of formation damage. On the other hand, the permeability ratio of 0.7 from *Rci* and *Rciii*, shows significant improvement, as earlier discussed. In addition, the porosity became greatly reduced (from 15.88 to 0.2 percent: *Rci* and *Rcii, respectively*) after wax deposition. However, there was a significant improvement to 8.35% after the treatment (*Rciii*).

Table 3—Petrophysical Parameters of Original Rock Sample *(Rci)*, Damaged rock sample *(Rcii)*, and Treated Rock Sample *(Rciii)* using the Optimum Conditions.

Samples	Permeability, *K* (mD)	Porosity, Ø (%)	Ratio of *K*	Ratio of Ø
Original (*Rci*)	0.01	15.88		
Damaged (*Rcii*)	0.0001	0.2	0.01	0.013
Treated (*Rciii*)	0.007	8.35	0.7	0.53

Conclusion

Thermochemical heat source has been introcuced as an advancement in varous stimulation technologies including formation damage restoration. Power ultrasound is an emerging green technology, which can enhance the performance of the conventional formation damage treatment technologies. The cleaning action of ultrasonication has to do with cavitations and implosion of bubbles, which is trasimmted as energy in the medium, which eventually assist in detaching contaminants from the surface. A facile applicability of the combined thermochemical treatment and ultrasonication has been demonstrated in this study. Ultimately, the current results revealed promising potentials for the proposed technology with scientific operational management, as supported in this study using empirical optimization approach.

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