

Functionalized Multiwall Carbon Nanotube Electrode for Electrochemical Oxidation of Dibenzothiophene in Diesel

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Abstract

Electrochemical oxidation techniques have attracted much attention in the last decade because they require moderate conditions of temperature and pressure in comparison to other processes that are used for desulfurization. An advanced electrochemical oxidation method to reduce sulfur content in diesel is proposed herein. In this work, functionalized multiwall carbon nanotubes (FMWCNTs) has been successfully employed for electrochemical oxidation of dibenzothiophene (DBT) in model diesel (n-Octane) using three electrode system. FMWCNTs was selected based on its low manufacturing cost and more often incorporating into large scale industrial applications. The influence of the main electrochemical oxidation parameters on the oxidation efficiency have been investigated. The electrochemical oxidation of DBT in n-octane was carried out in NaCl solution. Under the optimal conditions, the conversion of DBT in the model diesel reached 95.42% in 5 hours. n-Octane and NaCl solutions properties were examined before and after oxidation using Fourier transform infrared (FTIR) analysis which showed that the oxidation process has no impact on their properties.

Keywords: Multiwall carbon nano tube; Electrochemical oxidation; Dibenzothiophene; Cathode; Diesel; Electrolyte.

1. Introduction

Oxidation of sulfur compounds is a considerable interest for practical applications, specifically in the hydrodesulfurization (HDS) of petroleum distillates. Organic sulfur compounds in transportation fuels can be considered as the major source of air pollution, acid rain, and hazy weather due to the burning of high-sulfur-containing compounds [1]. However, a high temperature, high pressure, active catalysts, and large amount of hydrogen are necessary for HDS which requires a very high cost [2]. Current HDS techniques are efficient when they applied to aliphatic and cyclic sulfur compounds but are less effective for aromatic treatment [3].

To overcome these shortages of (HDS) and obtain a clean fuel, many alternative technologies have been developed in the recent years such as oxidation, extraction, adsorption, biodesulfurization, and alkylation [4-8]. Recently, oxidative desulfurization (ODS) has been considered as a promising alternative process to reduce sulfur in fuel [9-10]. Through the ODS process, sulfur compounds exist in liquid fuels can be converted into very polar sulfoxides or sulfones under mild conditions which can be easily extracted with polar solvent [11-12]. ODS has some advantages such as low operating cost, mild operating conditions, and easy technological process with the absence of hydrogen. In ODS process a variety oxidants can be used such as H₂O₂, O₂, and peroxy acid [13]. It can be seen in recent studies that the combination of oxidation and absorption or extraction can lead to a better desulfurization result [14]. However, the large amount of oxidizing material consumption and large amount of produced wastewater make it difficult to apply in the industrial application.

Electrochemical oxidative desulfurization (ECDS) of fuels has been proposed as a novel method of oxidative desulfurization in which no consumption of oxidizing agent and small

amount of wastewater is produced. In this process, the reactive oxidizing agent can be generated in situ by water electrolysis or based on electrolyte decomposition [15-17]. Electrochemical oxidation is an efficient method to in situ generate reactive oxidizing agent which attracts researchers as a new approach to liquid fuel desulfurization [18].

Schucker *et al.* [19-20] have invented a new method of electrochemical oxidation for removing sulfides from naphtha and the products of sulfide-oxide can be extracted easily by distillation. Wang *et al.* [21-22] developed a new electrochemical oxidation desulfurization method to remove organic sulfur compounds in gasoline in an electrochemical fluidized-bed reactor using β -PbO₂ load to the activated carbon surface as an anode. After electrochemical catalytic oxidation, the maximum desulfurization removal reached 87% by extraction. Li *et al.* [23] reported a process to reduce sulfur content in kerosene using NaCl solution as an electrolyte. Their data showed that 1-heptyl mercaptan can be oxidized effectively and the desulfurization efficiency reached 92.67%. Recently, Abdullah *et al.* [24] have proposed a new in situ oxidation technology for desulfurization of diesel in divided cells trickle bed electrochemical reactor (TBER) using carbon black. Their results showed that DBT can be oxidized in situ using TBER with conversion efficiency of 97.8%. More recently, Dávila *et al.* [25] reported an interesting study for electrochemical oxidation of dibenzothiophene compounds using BDD electrode in acetonitrile–water medium as an electrolyte. Their results suggested that DBT compounds can be oxidized in two pathways direct and indirect electrochemical oxidation reaction with hydroxyl radicals and other oxygen species that formed at BDD surface.

However, the development of ECDS technology is still in its early stage toward industrial applications and further researches are required to push this technology toward industrial applications. In this paper functionalized multiwall carbon nano tube (MWCNT) was used as a new approach for electrochemical oxidation desulfurization of DBT in diesel using three electrode systems. Linear sweep and cyclic voltammetry were used for electrochemical analysis and the influence of the main operation parameters (potential, electrolyte concentration, temperature and electrolyte to oil ratio) was systematically investigated to find the optimum conditions for DBT oxidation.

2. Experimental section

2.1. Materials

MWCNTs (outer diameter 15 ± 5 nm and length 15-20 μ m) were purchased from Nanolab Inc. (Newton, MA). PTFE aqueous suspension (60 wt.% dispersion in water), dibenzothiophene, and octane 98% were purchased from Sigma Aldrich (United States). H₂SO₄ (98%), KMnO₄, and HNO₃ aqueous solution were purchased from Alfa Aesar (United States). NaCl (AR, 99.5%) was obtained from Chengdu Kelong Chemical Co., Ltd.

2.2. Experimental methods

As shown in Figure 1, the electrochemical oxidation reaction experiments of diesel were conducted in an undivided electrolyte cell with different concentration of NaCl solutions. The cell consists of three electrode system in which functionalized MWCNTs was used as a working electrode, Ag/AgCl as a reference electrode and platinum wire as a counter electrode. MWCNTs electrode was prepared as follows: MWCNTs was functionalized with HNO₃ based on previous study [26], then 10gm of FMWCNTs was mixed with PTFE solution which works as a conductive binder. The mixture was then pasted on 2 x 5cm stainless steel mesh on both sides and dried at 80°C for 6h, and then calcined at 250°C for 2h. A 500 ppm of DBT was dissolved in 100 mL portion of n-Octane which was then filled into the cell and mixed with the prescribed NaCl solution (400 mL) under constant stirring (400 rpm) [18].

The electrochemical measurements were conducted using an Autolab PGSTAT 302 potentiostat/galvanostat from Eco Chemie, controlled by GPES4.9 software. After each experiment the oil was separated from the electrolyte and analyzed using gas chromatography (GC), Agilent 5973 Network Mass Selective Detector with an HP-5MS 5% phenyl methyl siloxane capillary column (30 m x 250 μ m x 0.25 μ m) and Fourier transform infrared (FTIR) spectroscopy

(Beijing Beifen-Ruili Analytical Instruments Co., Ltd.) were used to analyze DBT compound in model diesel and the electrolyte before and after oxidation.

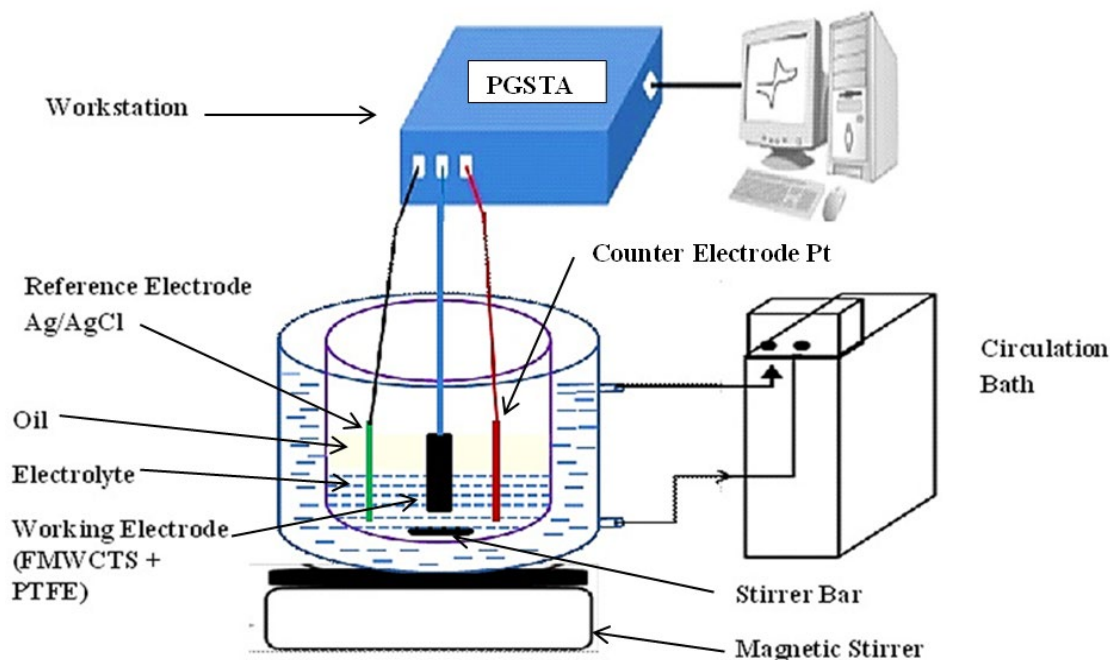


Figure1. Electrochemical oxidation experimental setup

3. Results and discussion

3.1. Electrochemical response of FMWCNTs electrode in NaCl solution

Cyclic voltammetry and linear sweep were reported in 0.1M NaCl electrolyte solution, mixture of NaCl solution with 10% n-Octane, mixture of NaCl solution with 10% n-Octane and DBT. Cyclic and linear sweep voltammetry for all above mentioned samples were conducted in the potential range of 0 to 1000 mV at 10 mV s^{-1} to clear observation of the oxidation process. As shown in Fig2a, the use of NaCl solution alone gives higher anodic current which could be attributed to the decomposition of NaCl and water to produce ClO^- which has ability to oxidize sulfur compounds [27], and might increase the electrical conductivity of the solution leads to higher current density. On the other hand, the presence of n-Octane in the electrolyte solution yields lower current density in comparison with free electrolyte. It is well known that n-Octane is nonconductive material which decreases the solution conductivity leading to decrease the current density of the mixture. It can be also observed that the presence of n-Octane and DBT together in NaCl solution shows the lowest current density. This phenomena can be explained as follows: the process herein is to oxidize DBT in n-Octane and as long as the increase in the current is the result of the formation of negative ions (ClO^-), it is expected that a certain percentage of these ions will go to the oxidation of the sulfur compound (DBT), which leads to a decrease in the current density. From Fig.2 b, it can be noted that the onset potentials of the NaCl solution, NaCl and n-Octane solution, and NaCl, n-Octane and DBT solution were 406 mV, 425 mV, and 30mV respectively. It is clear that in the range of 400-560 mV the oxidation curves were slightly increased for the electrolyte without and with n-Octane then increased sharply at potential higher than 560 mV, this is may be due to the high decomposition of NaCl at high potential. However, it can be noticed that the oxidation curve for the mixture with DBT started to increase slightly at very low potential, the reason of that may be due to the direct oxidation of DBT at the surface of electrode.

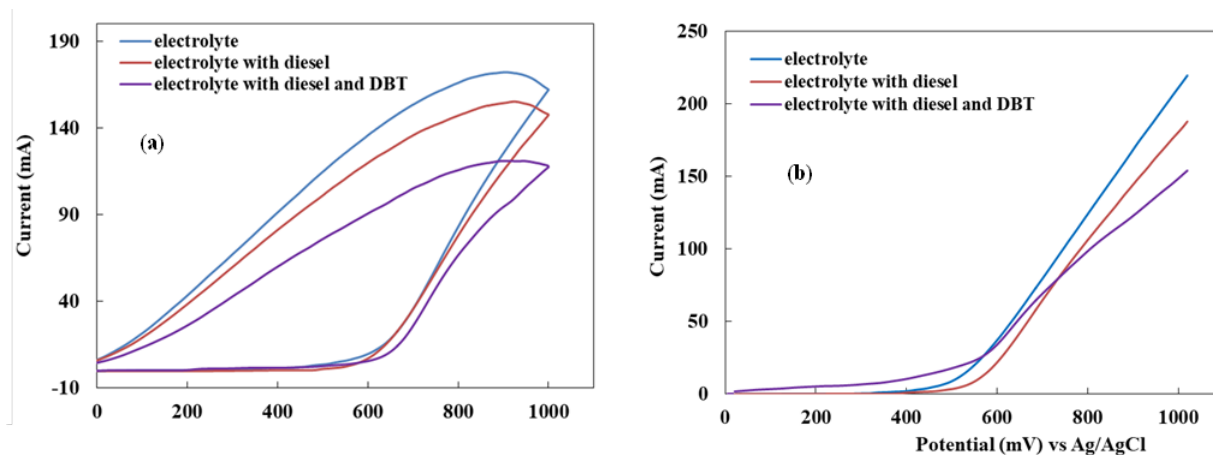


Figure 2. (a) Cyclic voltammetry Cv and (b) linear sweep voltammetry for electrolyte mixtures.

3.2. Impact of NaCl wt% on electrochemical oxidation of DBT in n-octane

The concentration of NaCl ranged from 5 to 30 wt%. NaCl concentration influence is presented in Figure 3. When salt is dissolved in water to form an electrolyte solution, it decomposes to form chloride ions Cl^- which can be oxidized by the supplied potential, a strong reactive oxidant (Cl_2 and ClO^-) can be produced which oxidize sulfur compounds to produce sulfone and sulfoxide [23,27]. These compounds have a high polarity which can be easily removed with polar solvent. Besides, Na^+ ions have no significant effect on oxidation efficiency as the slow raised of the oxidation efficiency was came from the increase of the electrical conductivity with the adding of NaCl [28].

As seen from Figure 3, the conversion of DBT increased initially and reached the maximum value at concentration of 20 wt% of NaCl, then decreased with increasing the weight percent of NaCl. The reason for this increase can be attributed to the fact that increasing the salt concentration leads to the provision of a greater amount of reactive ions Cl_2 and ClO^- and thus increases the oxidation process of the sulfur compound. However, increasing the electrolyte concentration to higher amount decreases the conversion. This may be attributed to the reduction in the produced reactive ions as a result of the above-normal concentrations as well as this will increase the viscosity of electrolyte solution, which might weaken the mass transfer and then finally leading to reduction in the DBT conversion.

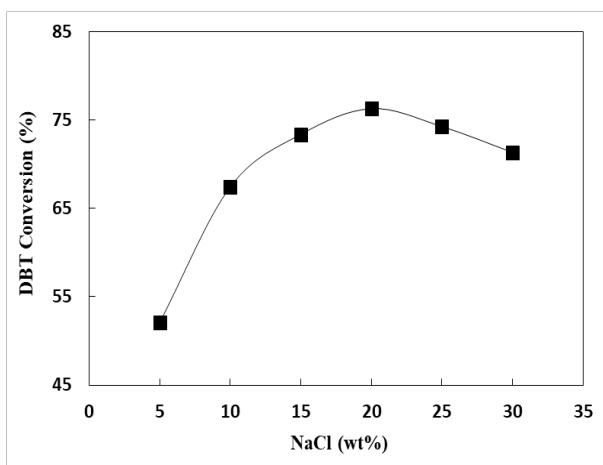


Figure 3. Effect of NaCl weight percent on electrochemical oxidation of DBT at 4 V, 20 °C, 60 min, volume ratio of electrolyte/oil, 1.0; and stirring rate, 500 rpm

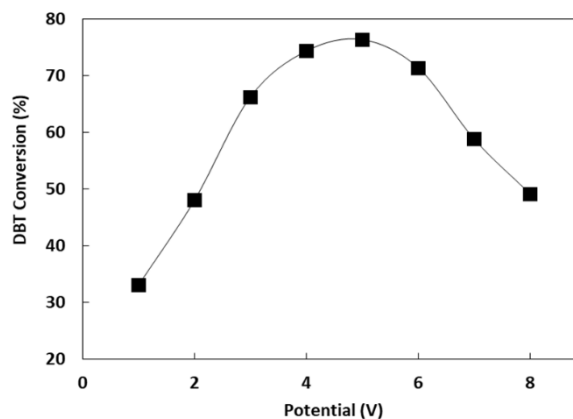


Figure 4. Effect of the applied voltage on electrochemical oxidation of DBT at 20°C, 60 min, volume ratio of electrolyte/oil, 1.0 and stirring rate of 500 rpm

3.3. Effect of applied potential

As shown in Figure 4, it is clear that the DBT conversion increases sharply with increasing the cell voltage and reach the maximum conversion at 5V, and then decreased at higher potential. It was reported that the applied potential has a significant influence on the electrode reactions [21]. Raising the applied potential might provide more energy to enhance the electrochemical oxidation reaction. However, excessive applied potential would lead to drop the current efficiency and power loss due to the side reaction such as oxygen evolution. Moreover, a high applied potential (>5V) could cause the oxidation of n-octane and change its color [23]. Therefore, an appropriate applied potential is critical factor. According to our experiments the optimal potential was 5V which is in a good agreement with previous study [23]. The generation of oxygen bubbles will separate the fuel from the electrode which inhibits the oxidation reaction. According to the first law of thermodynamics increasing the electrode over-potential caused by applied potential [29] will absolutely increase the rate constant resulting in speeds up the reaction rate [21].

3.4. Electrolysis temperature

The effect of temperature on DBT oxidation is presented in Figure 5. It can be observed that as the temperature increases, DBT conversion sharply increases and reaches the maximum oxidation efficiency at temperature of 45°C then decreases at high temperatures. Electrolysis temperature is a very significant factor in oxidative desulfurization process as it influences not only the reaction rate but also affect mass transfer rate [30]. Increasing the temperature could accelerate rate of the oxidation reaction. As long as the electrochemical oxidation reaction is exothermic reaction which released heat and raised the system temperature, the high temperature will make it difficult to support the reaction and restrained the oxidation reaction³¹. On the other hand, high temperature could accelerate the reaction of oxygen evolution from the supporting electrolyte which reduces the current efficiency resulting in loss of energy and effecting the oxidation reaction. Considering the economic and oxidation efficiency, 45°C was a suitable reaction temperature.

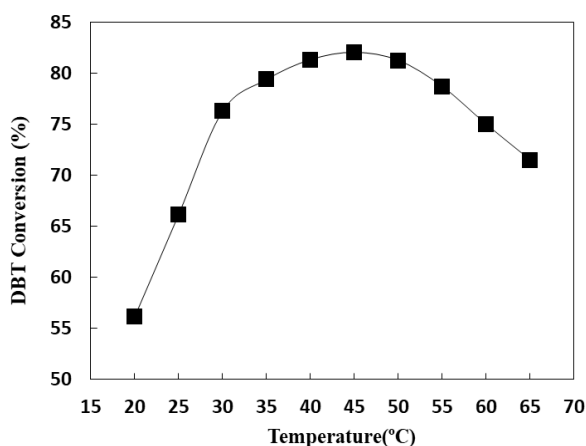


Figure 5. Effect of the electrolysis temperature on DBT electrochemical oxidation at 20 wt% of NaCl, applied potential 5 V, electrolysis time 60 min, volume ratio of electrolyte/oil 1.0 and stirring rate, 500 rpm

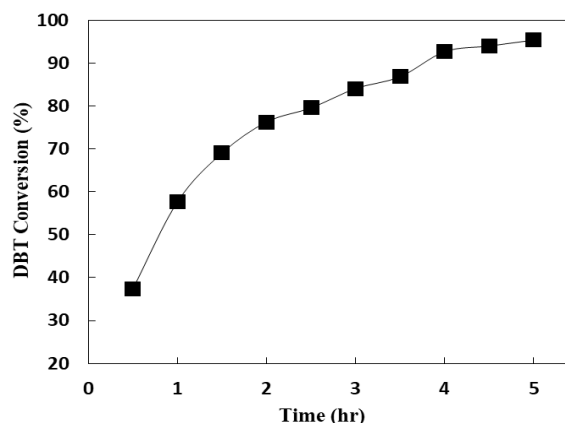


Figure 6. Impact of electrolysis time on DBT conversion at NaCl 20%wt, 45°C, 5V, volume ratio of electrolyte/oil, 1.0 and stirring rate of 500 rpm

3.5. Electrolysis time

It is another important technical factor for oxidation reaction of DBT and the effect of electrolysis time in 5h is shown in Figure 6. It reflects the oxidation reaction efficiency and determines the energy consumption of the experiments [32]. It is clear that DBT conversion rose

with the increase of electrolysis time. When the electrolysis time was less than 2h, prolongation the reaction time could efficiently enhance the oxidation reaction rate. However, when the time of electrolysis exceeds 2h, the increase in the oxidation reaction rate tend to be slow. Increasing the reaction time improves the production of reactive ions, Cl_2 and ClO^- which might continue to oxidize sulfur compound (DBT). Moreover, increasing the reaction time reduces the sulfur content to a very low level, so that extending the oxidation time might not enhance the oxidation reaction rate effectively.

3.6. Influence of electrolyte/oil volume ratio on DBT oxidation

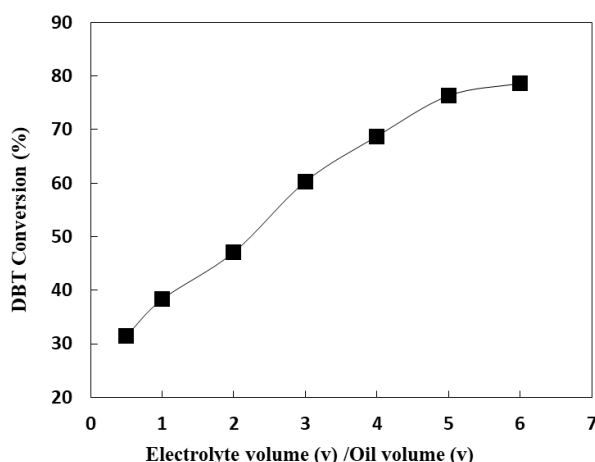


Figure 7. Influence of electrolyte/oil volume ratio on DBT conversion at NaCl (20 %wt), 45°C, 5 V, 1 h and 500 rpm

conversion remains almost the same. This could be attributed to the oxygen evolution as a result of too much electrolyte solution which might reduce the current efficiency and economic efficiency.

3.7. FTIR analysis for n-octane and NaCl electrolyte solution

The properties of n-octane and NaCl solution were investigated before and after oxidation using FTIR analysis. It is suggested that NaCl solution itself has no oxidation ability but the electrical current can pass through it resulting in oxidants production which play the main role in the oxidation process [23]. Figure 8 shows the FTIR spectra for the oil and the solution before and after electrochemical oxidation to give us some information for what happened to their properties.

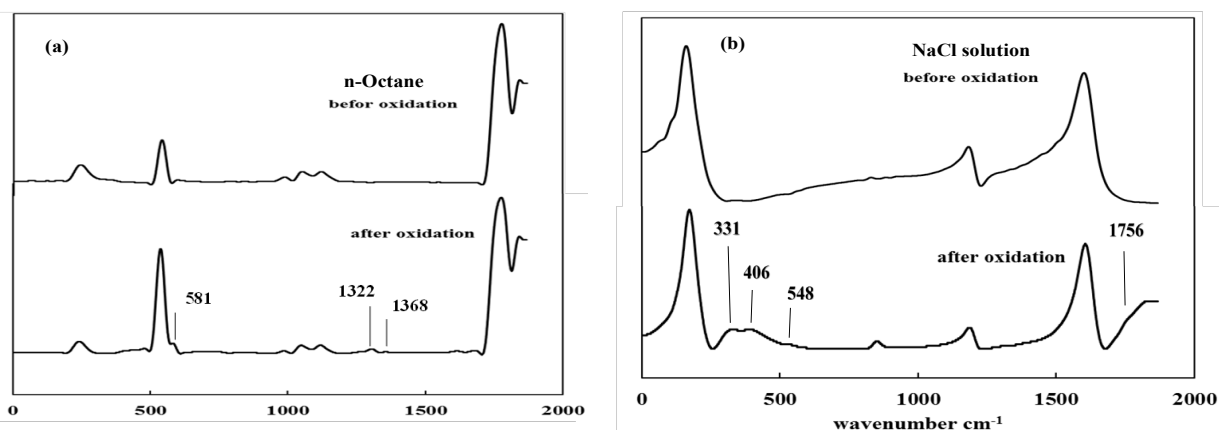


Figure 8. FTIR spectra for n-Octane and NaCl solution before and after electrochemical oxidation

Figure 8a illustrates the conversion of DBT in electrochemical oxidation technique. The peaks of 1368 cm^{-1} and 1322 cm^{-1} ($-\text{SO}_2$), and 581 cm^{-1} ($-\text{C-Cl}$) were observed in n-octane after electrochemical oxidation. The peaks 1322 and 1368 cm^{-1} represent the characteristic peaks of sulfoxide according to previous studies [34-35]. On the other hand, carbon-chlorine ($-\text{C-Cl}$) bond was also seen after electrochemical oxidation, this is might be due to the reaction of part of organic sulfides with ClO^- and Cl_2 to sulfonyl chloride compounds [23]. The presence of sulfoxide peaks confirm that DBT was transformed to the corresponding oxidation products which can easily extracted with polar solvent. The obtained spectra also revealed that there was no clear change in the available hydrocarbon phase. Figure 8b illustrates the FTIR spectra for NaCl solution before and after electrochemical oxidation. It can be noted that many peak existed at many different wave lengths of 331 , 406 , 548 and, 1756 cm^{-1} . These peaks maybe due to the electrochemical and/or chemical reactions during electrolysis of sodium chloride solutions which could produce several ions such as Cl^- , ClO^- , ClO_2^- , ClO_3^- which are beneficial for DBT oxidation.

4. Conclusion

The oxidation of DBT in n-octane by electrochemical oxidation in NaCl solution over FMWCNTs as a new electrode material in an advanced three electrode system has been investigated. Under optimal conditions the maximum oxidation conversion of 95.42% was achieved in 5 hours in which 500 ppm of DBT was reduced to 22.9 ppm. The results showed that DBT can be oxidized in this system and the selected electrode materials (FMWCNTs + PTFE) exhibited high efficiency in such electrochemical oxidation process. Finally, a possible electrochemical three electrode system for diesel oxidation desulfurization process has been proposed.

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