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Oxidation of dibenzothiophene to dibenzothiophene-sulfone using silica gel

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ABSTRACT

The oxidation of dibenzothiophene (DBT) to dibenzothiophene-sulfone (DBT-sulfone) has been a topic of particular interest in the past few years, as a possible technique to remove sulfur from crude oil. In this manuscript we show that the oxidation of DBT to DBT-sulfone occurs using silica and does not require the addition of an external oxidizing agent. The effect of the synthesis pH and the calcination temperature on the ability of the silica to oxidize DBT to DBT-sulfone was investigated. The oxidation reactions were performed by refluxing silica with DBT in the following solvents: decahydronaphthalene, tetrahydronaphthalene, dodecane, heptane, and toluene at different temperatures. The reaction was found to work only in hydrogen-donating solvents: tetralin and decalin. HPLC results show that 80% of the original 10,000 ppm DBT was oxidized when the reaction was carried out at 160 °C in decahydronaphthalene. FTIR was used to show that the product of the reaction was DBT-sulfone, which confirmed 80% conversion of the DBT to DBT-sulfone. The product of the oxidation reaction was further identified using powder XRD.

1. Introduction

Developed countries are setting new standards for the allowable amount of sulfur in diesel; in the United States, by 2014 sulfur levels in all diesel fuel must be below 15 ppm [1]. To meet these specifications at the pump, refiners must produce diesel containing one-half the amount of sulfur before it enters the distribution system, because the low-sulfur product is expected to pick up trace amounts of sulfur as it moves through pipelines and other distribution channels. Extra heavy oils, such as those from the Orinoco region in Venezuela or the Alberta tar sands in Canada, are typically upgraded in a process that is both capital- and energy-intensive.

Hydrodesulfurization processes are currently used to remove sulfur from crude oils. These processes use hydrogen which reacts with the sulfur compounds to generate H_2S gas. These processes involve high temperatures of about 350 °C and high pressures of H_2 gas from 3 to 5 MPa [2]. From an economic and environmental point of view, it would be more suitable if low-temperature and low-pressure systems could be developed to remove sulfur from fuels. High-temperature and high-pressure processes decrease the lifetime of the catalyst and consumes larger amounts of H_2, which results in higher costs. In addition, high-temperature and high-pressure processes result in the generation of H_2S, a highly toxic compound. Another important reason to find an alternative method to remove sulfur is that substituted sulfur compounds such as methyl ethyl dibenzothiophene, 4-methyl dibenzothiophene, 3 methyl dibenzothiophene, and others are non-reactive under hydrodesulfurization process [3–8].

An alternative method to remove sulfur from fuels is oxidative desulfurization. There have been many catalysts investigated to oxidize sulfur-containing molecules in organic solvents. For example, Mo/Al_2O_3 catalyst has been studied for the oxidative desulfurization (OD) process in conjunction with hydrogen peroxide as the oxidizing agent [9]. Another example of an oxidation desulfurization catalyst is $Ti_3(PW_{12}O_{40})_4$, which also requires the use of H_2O_2 as an oxidizing agent [10]. Potassium superoxide has shown sulfur removal as high as 99% with samples of benzothiophene, dibenzothiophene, and a number of selected diesel oil samples [11].

As mentioned before these oxidative desulfurization techniques utilize oxidizing agents such as NO_2, H_2O_2, and *tert*-butyl-hydroperoxide. The advantage of using these techniques is that sulfur can be removed at relatively low temperatures and atmospheric pressure. The major drawback to ODS is that current catalytic systems require the use of an oxidizing agent such as H_2O_2. The constant use of strong oxidizing agents such as hydrogen peroxide can become expensive and makes these catalysts cost prohibitive on the large scale.

In the present study, the oxidative catalytic properties of SiO_2 were investigated. First, silica gel grade 22 was obtained from Alfa Aesar and tested in the dibenzothiophene (DBT) model reaction. Then, silica gel was synthesized at different pHs and tested

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in the model reaction. Fourier-Transformed Infrared spectroscopy was used to follow the oxidation of DBT (dibenzothiophene) to DBT-sulfone at the different pHs and different reaction temperatures. X-ray diffraction was used to characterize the catalysts before and after the reactions. In addition, X-ray diffraction was used to confirm that the product was DBT-sulfone. HPLC (high performance liquid chromatography) was used to determine the percentage removal of DBT after each reaction.

2. Experimental

2.1. Synthesis of silica gel at different pH

Silica was synthesized at different pHs. Silica gel was prepared with slight modifications to previously reported methodologies [12–15]. Sodium metasilicate (Na_2SiO_3) and hydrochloric acid (HCl) were obtained from Alfa Aesar. Na_2SiO_3 was dissolved in water, which produced a basic solution (between pH 12 and 14). HCl was added to the solution under vigorous stirring to reduce the pH to 10, which resulted in a gel. Subsequent to formation the gel was then washed several times with water to remove any NaCl and other byproducts of the synthesis. In addition, the gel was then washed with alcohol and filtered under vacuum to remove any residual water. The resulting gel was then dried at three temperatures: 100 °C (24 h), 140 °C (24 h), and 380 °C (24 h). Similarly, SiO_2 gels were synthesized at pHs 9, 8, and 7 using the same precipitation procedure, washing, and drying conditions. The synthesis of SiO_2gel at pHs 6, 5, and 4 consisted of bringing the pH down to approximately 2 and subsequently raising the pH to either 4, 5, or 6 using sodium hydroxide. SiO_2 was also synthesized at very acidic pHs 3, 2, and 1. The acidic synthesis technique was similar as previously mentioned. HCl was added quickly to a solution of Na_2SiO_3 in water under vigorous stirring to achieve a pH of 1. The solution was then heated to form a gel, which was subsequently washed with water and alcohol and dried as mentioned earlier.

2.2. DBT oxidation reaction

The oxidative properties of SiO_2 synthesized at different pHs were tested using the DBT model reaction. DBT (0.5 g) was dissolved in decalin (50 mL), 0.5 g of SiO_2 was added, and the reaction was refluxed for 4 h under vigorous stirring. The reactions were tested at six different temperatures: 50 °C, 100 °C, 120 °C, 140 °C, and 160 °C. Different temperatures were tested to determine the optimum temperature of the reaction. In addition, different solvents were tested which included decalin, tetralin, toluene, heptane, and dodecane. Different solvents were used to determine if the solvent and the temperature are important factors controlling the oxidation of DBT to DBT-sulfone.

2.3. High performance liquid chromatography (HPLC)

High performance liquid chromatography (HPLC) was performed with a SPHERI-5 (5 μm) Silica-based column, a Spectra-Physics Spectra System P1500 gradient pump, UV2000 detector, and Winner for Windows Software. The mobile phase will be 95% hexanes, 5% isopropanol by volume with helium solvent de-gassing. The flow rate was set to 1 mL/min. and the detector was set to a 256-nm wavelength, all samples were run at room temperature. A 1 μl aliquot of analyte was used and in some samples it was necessary to increase the amount to enhance the signal. A calibration curve was obtained by using four different concentrations of DBT. The correlation coefficient of the calibration curves used in this study was $R^2 = 0.99$ or better.

2.4. FT-IR infrared spectroscopy

Infrared spectroscopy was carried out with a Thermo Nicolet Nexus 470 FT-IR. The samples for IR measurements were prepared as pellets by embedding the sample in a polycrystalline KBr matrix. The pressure applied to make the pellets was 5 MPa. In addition, a background spectrum was collected before each sample spectrum collection.

2.5. X-ray diffraction (XRD)

The X-ray diffraction data were collected using a D5000 X-ray diffractometer with Cu Kα radiation with a wavelength corresponding to 1.540 Å. The start angle was 5° and the stop angle was 45°.

3. Results

3.1. High performance liquid chromatography results

SiO_2 synthesized at different pHs in the range 3–10 showed no reactivity toward the DBT. However, when SiO_2 was synthesized in the pH range pH 1–2 the oxidation of DBT to DBT-sulfone was observed. In addition, it was also observed that the percentage of DBT removal increased as the calcination temperature of the gel was increased from 100 °C to 380 °C as shown in Fig. 1A. As it can be seen in Fig. 1A when the gel is dried at 100 °C only a 20% reduction of the DBT concentration was observed. The concentration of DBT removed when the SiO_2 was calcinated at 140 °C was 40%. However, at a calcination temperature of 380 °C (above the critical point of water), a reduction of 80% of the DBT concentration was observed. A comparison between commercially available silica and the in-house-synthesized silica for the oxidation of DBT was performed. The reactions of both the in-house-synthesized silica and the commercial silica with DBT were performed at 100 °C, 140 °C, and 160 °C and are shown in Fig. 1B. From the three temperatures used 100 °C, 140 °C and 160 °C, the reaction run at 160 °C was the one that showed the largest removal of DBT. Furthermore, it was noted, as shown in Table 1, that the oxidation of DBT to DBT-sulfone occurs only in the presence of a hydrogen-donating solvent such as decalin or tetralin. No oxidation was observed when non-hydrogen donor solvents such as heptane and toluene were used. The conversion using either tetralin or decalin is approximately the same. For simplicity, only data using decalin as a solvent are shown in Fig. 1B. According to the data shown in Fig. 1B, the highest removal, 85%, of DBT occurred at 160 °C. Finally, Fig. 1C shows the time requirements for the removal of DBT at 160 °C, using sampling times of 60, 120, and 240 min: the reaction requires 240 min to reach 80–85% removal of DBT.

3.2. FT-IR results

The results of the reaction using DBT as the model compound are shown in Fig. 2. These results correspond to the products of the reactions carried out at 160 °C using silica grade 22 and the laboratory-synthesized silica (pH 1) as catalysts (Fig. 2A and B). Fig. 2C and D shows the infrared spectra corresponding to DBT-sulfone standard and DBT, respectively.

In Fig. 3, the infrared spectrum is shown only from 800 to 400 cm^{-1} wavenumbers where the C–S and S–O ring vibrations occur. It is important to emphasize the difference in the infrared spectrum after the oxidation reaction using silica at 160 °C (Fig. 3A). The shift in the peak at 744 cm^{-1} occurs in the treated samples and the absorption bands corresponding to S–O vibrations appear in the 600–500 cm^{-1} range (Fig. 3A). Comparing the treated

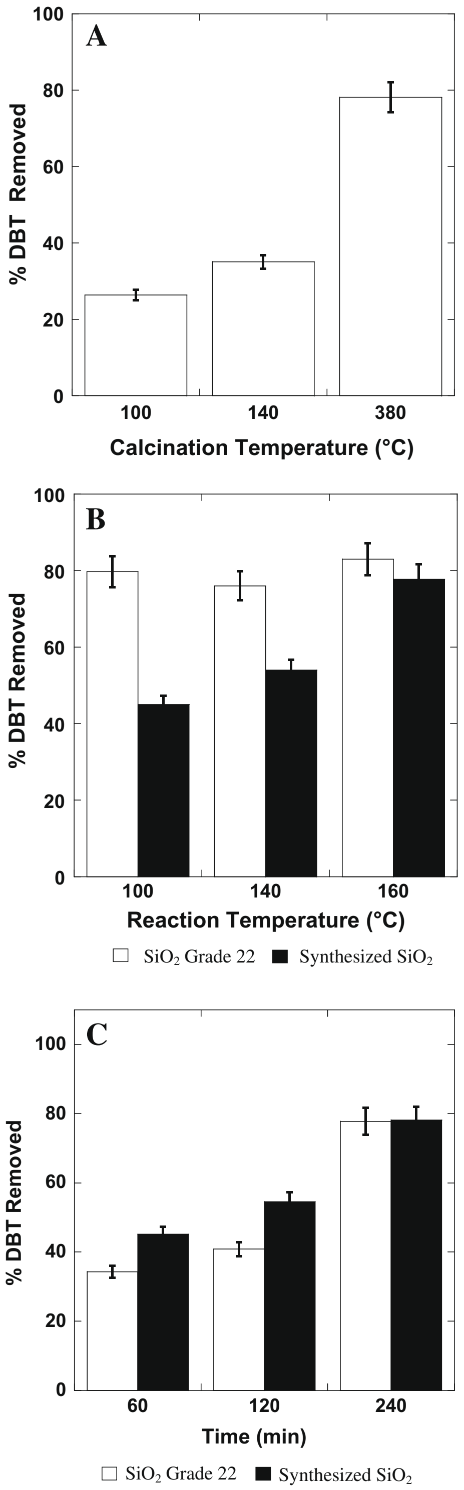

Fig. 1. (A) Effect of calcination temperature of acidic SiO_2 on the removal of DBT from a decalin solution at 160 °C. (B) Effect of reaction temperature on the removal of DBT in the presence of SiO_2 grade 22 and synthesized acidic SiO_2 calcinated at 380 °C. (C) Comparison of time of the removal of DBT using SiO_2 grade 22 and synthesized acidic SiO_2 (calcinated at 380 °C) at a reaction temperature of 160 °C.

sample to DBT standard (Fig. 3D) there is only a fraction of DBT left in the sample (approximately 20%). Comparing the treated sample to DBT-sulfone standard (Fig. 3C), all the vibrations of the DBT-sulfone standard are present in the treated samples.

Table 1
Results of reactions of DBT with different SiO_2: commercially available SiO_2 grade 22, SiO_2 basic, SiO_2 neutral, and SiO_2 acid in house synthesis, using different solvents, and reaction temperatures.

Catalyst	Reaction temperature (°C)	Solvent	%DBT removal
SiO_2 grade 22	160	Decalin	80
SiO_2 grade 22	160	Tetralin	85
SiO_2 grade 22	100	Heptane	0
SiO_2 grade 22	110	Toluene	0
SiO_2 grade 22	160	Dodecane	0
SiO_2 neutral pH	160	Decalin	0
SiO_2 basic pH	160	Decalin	0
SiO_2 acidic pH	160	Decalin	80
SiO_2 neutral pH	160	Tetralin	0
SiO_2 basic pH	160	Tetralin	0
SiO_2 acidic pH	160	Tetralin	84

Fig. 4 shows DBT after reaction with laboratory-synthesized silica calcinated at different temperatures: 100 °C, 160 °C, and 380 °C. Fig. 4A and B shows DBT after reaction with laboratory-synthesized silica calcinated at 380 °C and 160 °C, respectively. Fig. 4C shows DBT after reaction with laboratory-synthesized silica calcinated at 100 °C. Absorption bands of Fig. 4A and B coincide with the absorption bands of DBT-sulfone standard (refer to Fig. 3D); however, when the laboratory-synthesized silica is calcinated at 100 °C, conversion of DBT to DBT-sulfone is not clearly shown. As can be seen in Fig. 4 the appearance of the DBT-sulfone vibrations becomes more defined as the calcination temperature of the silica increases.

The main differences that were observed when the reaction is run at different temperatures are in the range of 1000–1500 cm^{-1}. The absorption bands corresponding to DBT-sulfone start appearing when the reaction is run at 100 °C (data not shown). However, DBT-sulfone vibrations between 1000 cm^{-1} and 1500 cm^{-1} are masked by the presence of the silica, which absorbs in the range of 1000–1500 cm^{-1} and thus not visible in the FT-IR spectra. As the reaction temperature increased, the absorption bands of DBT-sulfone standard in the range of 1000–1500 cm^{-1}, began to become visible in the samples treated with SiO_2. Based on these data, it is suggested that a lower temperature DBT is absorbed onto the silica first and is subsequently oxidized. As the temperature at which the reaction is run increases (160 °C), the silica releases the oxidized DBT (DBT-sulfone) back to the solution. The oxidation reaction was studied at the boiling temperature of the decalin (190 °C). At boiling point of the decalin no oxidation was observed, not even partial oxidation was observed (data not shown) (see Fig. 5).

3.3. XRD results

The X-ray diffraction patterns of DBT-sulfone obtained and of DBT-sulfone standard are shown in Fig. 6. Cerius software was used to assign the *h k l* planes. DBT-sulfone was modeled using the cell parameters and atomic positions as previously reported [16]. The unit cell parameters used were $a = 10.09$ Å, $b = 13.89$ Å, $c = 7.22$ Å, and $\beta = 93.5°$. The space group used was C2/c. The X-ray diffraction patterns showed a difference in the intensities (Fig. 5). Reflections at 13.30 and 22.14 2θ correspond to the 020, and 220 planes and are in the same intensity in DBT-sulfone standard and SiO_2-reacted DBT. The treated sample may have adopted preferred orientation in the 110, $\bar{1}11$, 021, 040, 221, 041, and $\bar{1}13$ planes since they are the predominant peaks in the treated sample. It is possible that the height peaks are different because the treated sample was more crystalline than the purchased sulfone. The way the treated sample was extracted from the solvent was by slow evaporation which led to the formation of large DBT-sulfone crystals.

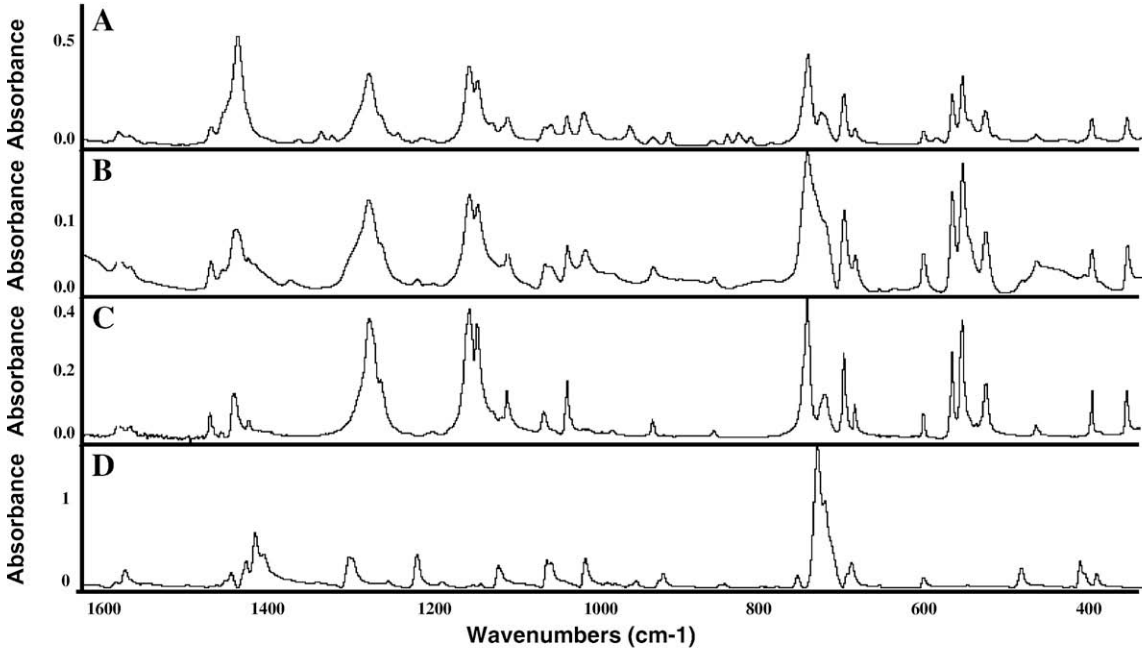

Fig. 2. Mid-IR of DBT after reaction with silica grade 22 and laboratory-synthesized silica. DBT after reaction with silica grade 22 run at 160 °C (A). DBT after reaction with laboratory-synthesized silica at pH 1 (B). DBT-sulfone standard (C). DBT standard (D).

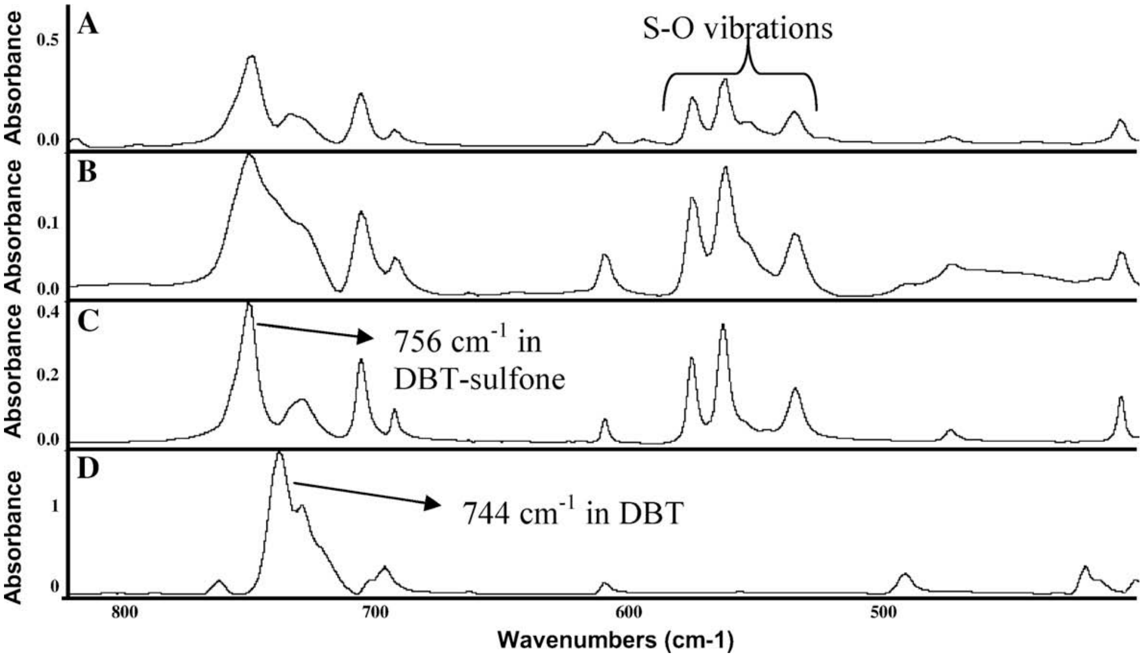

Fig. 3. Far IR of DBT after reaction with silica gel grade 22 at 160 °C (A). DBT after reaction with laboratory-synthesized silica at pH 1 (B). DBT-sulfone standard (C). DBT standard (D).

Only when the reaction was run at higher temperatures at 160 °C DBT-sulfone crystals could be extracted and observed in the X-ray diffraction pattern. When the reaction was run at a lower temperature between 100 and 140 °C DBT-sulfone could not be completely isolated and the X-ray diffraction pattern showed silica (a broad diffraction peak at about 20 in 2θ) and DBT-sulfone diffraction peaks.

In addition, silica gel grade 22 and the acidified silica synthesized and calcinated at 100 °C, 160 °C, and 380 °C were also characterized using X-ray diffraction as shown in Fig. 6 (for simplicity only the silica synthesized at 100 °C is shown). X-ray diffraction results showed that silica grade 22 as well as silica calcinated at the different temperatures were amorphous, showing only a broad weak diffraction peak centered around 20 ° in 2θ.

4. Discussion

Table 1 shows a summary of the results of the reactions run using silica and different solvents. As can be seen in Table 1, both the reaction temperature and solvent directly affect the oxidation

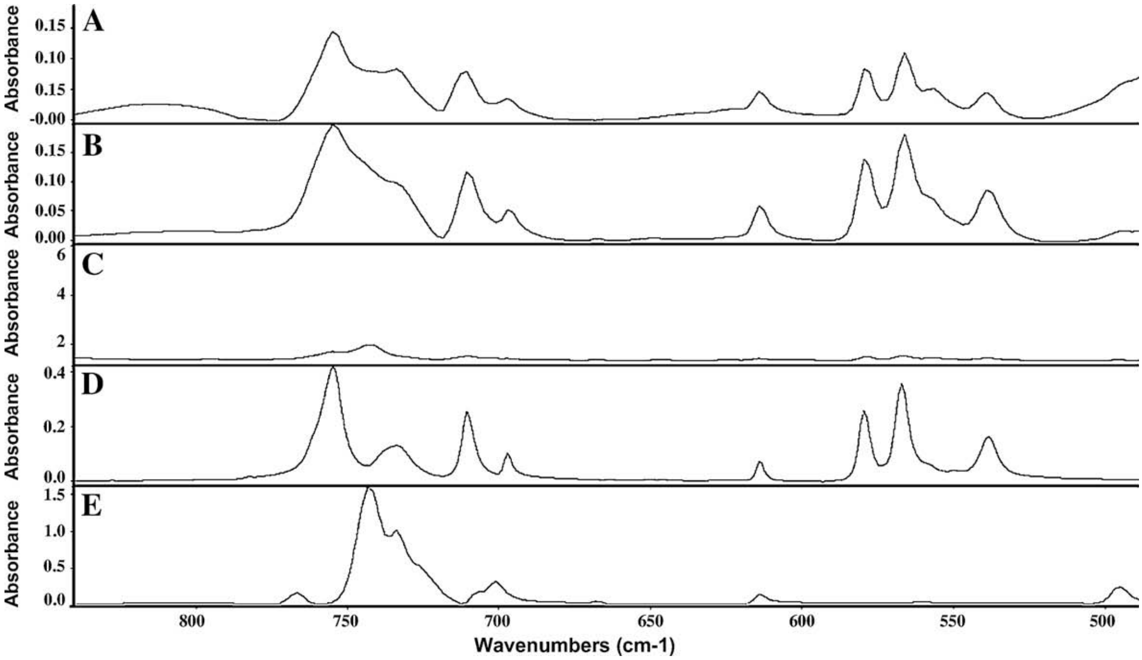

Fig. 4. Far-IR of DBT after reaction with laboratory-synthesized SiO_2 calcinated at different temperatures. DBT after laboratory-synthesized silica calcinated at 380 °C (A). DBT after laboratory-synthesized silica calcinated at 160 °C (B). DBT after laboratory-synthesized silica calcinated at 100 °C (C). DBT-sulfone standard (D). DBT standard (E).

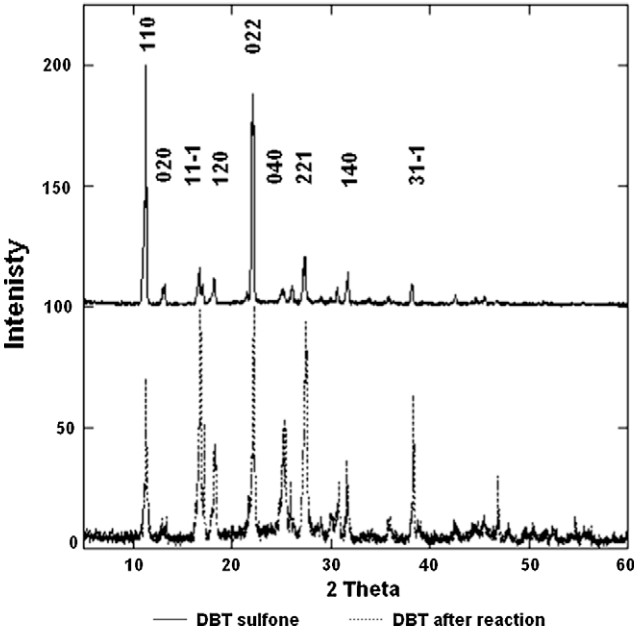

Fig. 5. X-ray diffraction pattern of DBT-sulfone standard (dotted line) and DBT after reaction with silica gel grade 22 at 160 °C (solid line).

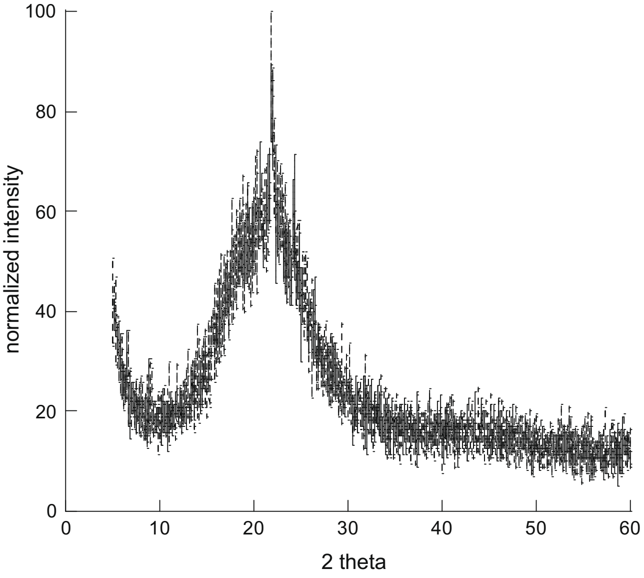

Fig. 6. X-ray diffraction pattern of SiO_2 synthesized at 100 °C.

of DBT to DBT-sulfone. The oxidation reaction was also carried out in tetralin (1,2,3,4 tetrahydronaphthalene) at the same temperatures at which the decalin reaction was carried out, 100 °C, 120 °C, and 160 °C. The reactions carried out in tetralin had the same behavior as the reactions carried out in decalin. At lower temperatures, the DBT-sulfone was stuck on the silica and at higher temperatures was released back to the solution. At the boiling temperature of tetralin and decalin, oxidation did not occur. This suggests that dissolved gases (such as O_2) in the solvent may be partially responsible for the oxidation; as the temperature increases the solubility of the gases decreases. At the boiling point of the solvent all the dissolved gases are released and thus there is no gas left in the solution for the conversion of DBT to DBT-sulfone. In addition, the reactions were also studied in non-hydrogen-donating solvents: heptane, dodecane and toluene at different temperatures. The oxidation of DBT to DBT-sulfone did not occur in either of these solvents at any tested temperature. This result suggests that in addition to needing a dissolved gas in the solvent, a hydrogen-donating molecule is also necessary to oxidize DBT.

In addition to the reaction temperature and solvent type, the pH of the SiO_2 synthesized plays an important role in the oxidation reaction. SiO_2 (silica gel) synthesized in the pH range of 3–10 had no observable reaction with the DBT molecule. The oxidation of DBT occurred only with SiO_2 synthesized at a pH between 1 and 2. The effect of the synthesis pH on the reaction may be linked to the structure of the SiO_2, which is dependent on the synthesis pH of the SiO_2. A SiO_2 synthesized at acidic pHs will have H^+ embedded in the matrix, whereas a silica gel synthesized under basic pH will have OH^- embedded in the matrix [17]. Synthesis under acidic pH conditions causes condensation to occur mainly between the neutral silanol groups (Si–OH) and protonated silanol groups ($Si–OH^+$), which are monomers and at the ends of polymeric chains of SiOH lead to the formation of linear polymers [17]. However, the synthesis under basic pH conditions causes the condensation of Si–OH groups through reaction of a deprotonated silanol ($Si–O^-$), in the middle of the chains, and a neutral silanol (Si–OH) or Si–OR group at the end of chains [17]. Comparatively, the acid-catalyzed or acidic SiO_2 compounds contain higher concentrations of adsorbed water, silanol groups, and unreacted alkoxy (depending on the source of the Si in the synthesis) groups than base-catalyzed precipitates [17]. From the results of the current study the oxidation of DBT occurs only in the presence of acidic silica, which suggests that the $SiOH^+$ groups are important in the reaction mechanism. It is hypothesized that the H^+ protons from the acidic SiO_2 are the necessary driving force for the oxidation reaction. It has been reported that sulfides can be oxidized to sulfoxides using HNO_3 supported on silica gel and polyvinylpyrrolidone (PVP) [18]. The oxidation of DBT to DBT-sulfone only in the presence of acidic SiO_2 suggest that the reaction is at least partially acid catalyzed, however, a hydrogen-donating solvent is necessary for the reaction to occur.

Furthermore, it was also observed that the calcination temperature of the SiO_2 has an impact on the oxidation of DBT to DBT-sulfone. Silica gel calcinated at 380 °C had the highest percentage conversion of DBT to DBT-sulfone.

Further studies are necessary to determine the reactions mechanism of the oxidation of DBT using silica. The literature shows the use of SiO_2 in conjunction with peroxides and other oxidizing solvents such as aldehydes and carboxylic acids to oxidize DBT to DBT sulfone [19–27]. In the current study it is being hypothesized that the in situ production of H_2O_2 is responsible for the oxidation of DBT to DBT-sulfone. The combination of the acidic SiO_2, dissolved gases in the solvent, and the presence of the hydrogen-donating solvent could very easily produce H_2O_2 or OH radicals in situ; however, further study is needed to clarify the reaction mechanism.

5. Conclusion

The current study addressed the use of acidified silica gel for the oxidation of DBT in the presence of a hydrogen donor solvent, at relatively low temperature and atmospheric pressure, without the addition of an oxidizing agent. Amorphous acidified silica (synthesized at an acidic pH) is capable of acting as an oxidizing agent in the oxidation of DBT to DBT-sulfone in the presence of a hydrogen donor solvent. In addition, the reaction occurs over a temperature range of 100–160 °C with a maximum removal, 80–85%, of the DBT occurring at 160 °C. According to the results obtained in this study sulfur compounds in heavy feeds could be oxidized and removed using a combination of acidified silica and a hydrogen-donating solvent. Further studies are needed to determine the mechanism of this reaction if it is either acid catalyzed or H_2O_2 is being made in situ.

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