

# Original Article: *Different Weight Percentages Doubled in the Catalyst*

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## ABSTRACT

In order to optimize the amount of dopant, 3 catalysts with different weight percentages of dopant entered the test medium and it was found that the increase of silver, cerium, nickel. It is one of the factors that accelerates the process of destruction. But this increase works to some extent, and more than this amount will have the opposite effect. In order to optimize the amount of dopant, the oxidative activity of the di benzothiophene compound in a simulated mixture of petroleum derivatives on a set of cerium catalysts as well as nickel deposited on a titanium dioxide substrate was doped with various amounts of Ni dopant. In general, the oxidant enters the test medium to facilitate degradation. All experiments with the presence of oxidant had a higher efficiency. Therefore, in order to optimize the amount of oxidant, different amounts of them were introduced into the test medium and it was found that increasing the oxidant is one of the factors that accelerates the degradation process, but this increase is somewhat effective and more destructive.

## Introduction

Strict environmental laws around the world to reduce environmental pollution have drawn researchers' attention to the deep desulfurization of fuels used in

transportation. The American Environmental Protection Agency enacted laws that forced the refining industry to reduce the sulfur content of gasoline and diesel fuel to about 30 ppmw and 15 ppmw in 2006 [1-3].

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**Table 1:** The effect of different doped weight percentages on efficiency

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst type
0%					<b>Pcat(17)</b> <b>Ce(%32)/TiO<sub>2</sub></b>
33.52%	4 hours	Visible	-35% ml0.2	0.5gr	<b>Pcat(18)</b> <b>Ce(%16)/TiO<sub>2</sub></b>
35.65%					<b>Pcat(19)</b> <b>Ce(%8)/TiO<sub>2</sub></b>
99.40%					<b>Pcat(21)</b> <b>Ni(%8)/TiO<sub>2</sub></b>

Because excess H<sub>2</sub>O<sub>2</sub> in its environment can act as a radical trap for OH • and reduce the overall rate of degradation. Therefore, a controlled increase in H<sub>2</sub>O<sub>2</sub> improves the reactivity of oxidative desulfurization (ODS) in sulfur

compounds, because the excess oxidants increase the equilibrium reaction and the thermal decomposition of the oxidant in the reaction reacts to the oxidation reactions [4-7].

**Table 2:** Comparison of the results obtained by changing the amount of auxiliary oxidant in the amount of efficiency

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	mass (gr)	Catalyst type
5.81%			-		
2.03%	4hours	UV-C	-35% ml 0.8	0.07gr	<b>TiO<sub>2</sub>(P25)</b>
27.06%			-35% ml0.5		
32.60%			-35% ml 0.2		

With increasing hydrogen peroxide, the process efficiency increases, reaches a maximum at one point and decreases with increasing H<sub>2</sub>O<sub>2</sub>. These inconsistencies are explained by the fact that since H<sub>2</sub>O<sub>2</sub> is a strong oxidant as well as a good receptor for electrons, it can trap electrons in the electron-hole pair produced by optical excitation and prevent the leaf from hatching. Catalyze at the surface [8-11]. Hydrogen peroxide acts as an electron acceptor like oxygen and can compensate for the lack of oxygen. H<sub>2</sub>O<sub>2</sub> is also refracted under UV light to produce

hydroxyl (OH) radicals, which are useful for the photocatalytic process [12-15]. Therefore, during the two described processes, the number of hydroxyl radicals produced in the photocatalytic reaction and the resulting degradation efficiency can be directly attributed to the amount of H<sub>2</sub>O<sub>2</sub>. On the other hand, H<sub>2</sub>O<sub>2</sub> itself can play the role of a radical trap (OH •). Excess H<sub>2</sub>O<sub>2</sub> is usually recombined with free hydroxyl radicals and reduces the overall rate of degradation [16-19].

**Table 3:** Comparison of the results obtained by changing the irradiation time in the efficiency

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst type
45.80%	2hours			0.3	
49.24%	3hours			0.3	
54.74%	4hours			0.3	
76.60%	6hours	UV-C	-35%ml0.2	0.3	<b>TiO<sub>2</sub>(P25)</b>
98.03%	2hours			0.7	
98.90%	3hours			0.7	
99.69%	4hours			0.7	

### Type of light radiation

In the batch photo reactor designed in this research, the experiments were subjected to two types of UV-C and Visible radiation. UV-C (190-

250 nm) ultraviolet rays carry a risk of cancer due to their short wavelength. While the wavelength of the visible spectrum is between 380-750 nm, it will not pose any danger to the environment [20-23].

**Table 4:** Comparison of the results obtained from the type of light radiation in the amount of efficiency

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst type
0%		UV-C			<b>Pcat(7) Ag(%8)/TiO<sub>2</sub>(P25)</b>
7.45%		Visible			<b>Pcat(7) Ag(%8)/TiO<sub>2</sub>(P25)</b>
9.63%	4hours	UV-C	-35%ml0.2	0.5gr	<b>Pcat(16) Ag(%8)/TiO<sub>2</sub>(TTIP)</b>
17.09%		Visible			<b>Pcat(16) Ag(%8)/TiO<sub>2</sub>(TTIP)</b>
23.49%		UV-C			<b>Pcat(11) Ce(%8)/TiO<sub>2</sub>/NaX</b>
99.45%		Visible			<b>Pcat(11) Ce(%8)/TiO<sub>2</sub>/NaX</b>

**Table 5:** Comparison of the results of changing the efficiency of desulfurization process by increasing 10 times the volume of primary feed

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst type	Primary feed volume
10.45%			2ml -35%	0.7		
77.17%		UV-C	0.2ml -35%	2	TiO <sub>2</sub> (P25)	<b>(ml)100</b>
79.09%	4hours		-35%ml1	2.5		

**Table 6:** Comparison of the results of changing the efficiency of desulfurization process by doubling the initial feed volume

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst type	Primary feed volume
100%	4hours	UV-C	-35%ml 0.4	1.4gr	TiO <sub>2</sub> (P25)	<b>(ml)20</b>

### Photo catalyst type

In this study, to perform the oxidative desulfurization process, 30 different Nano photo catalysts according to table (7), designed and constructed with differences in the percentage

of weighted loading or doping, differences in the loaded or doped component in the base, number of components involved in loading or doping (including two-component loading or doping) and differences in the type of bases doping has taken place in them. It should be noted that the

difference between loading and doping catalysts according to the results of their characterization analyzes is due to differences in the method of fabrication of these photo catalysts [24-27]. In

loading catalysts, the loaded component is placed on the base surface, while in doping catalysts, the desired dopant enters the base crystal structure [28-31].

**Table 7:** List of synthesized photo catalysts with high degradation efficiency

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst conditions	Catalyst type
99.45%					8% cerium loaded on TiO <sub>2</sub> and NaX zeolite	<b>Pcat(11)</b>
99.40%	4hours	Visible	-35% ml 0.2	0.5gr	8% doped nickel in TiO <sub>2</sub> (with TTIP precursor)	<b>Pcat(21)</b>
99.97%					8% cerium doped in TiO <sub>2</sub> and then doped in NaX zeolite	<b>Pcat(28)</b>
99.99%					8% nickel doped in TiO <sub>2</sub> and then doped in NaX zeolite	<b>Pcat(29)</b>

**Table 8:** Comparison of the efficiency of group A photo catalyst in desulfurization

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst type
5.58%				0.3	<b>Pcat (1)</b>
17.63%				0.7	
35.98%				0.5	
3.08%				0.5	<b>Pcat (2)</b>
8.93%				0.1	
12.45%	4hours	UV-C	-35%ml 0.2	0.3	
11.43%				0.7	<b>Pcat (3)</b>
37.05%				0.5	
32.28%				0.3	
49.03%				0.7	<b>Pcat (4)</b>
50.35%				0.5	
21.61%				0.5	
10.95%				0.3	<b>Pcat (5)</b>
23.70%				0.5	

**Table 9:** Relationship between TiO<sub>2</sub> (P25) loaded in photo catalyst and DBT reduction percentage

Efficiency	Percentage of TiO <sub>2</sub> (P25) loaded on the photo catalyst	Catalyst type
35.98%	55.46%	<b>Pcat(1)</b>
37.05%	43.37%	<b>Pcat(2)</b>
50.35%	31.68%	<b>Pcat(3)</b>
21%	22.62%	<b>Pcat(4)</b>
23.70%	15%	<b>Pcat(5)</b>

To justify this, it can be said that when TiO<sub>2</sub> (P25) is used alone as a photo catalyst, it works better in larger quantities, but when loaded or doped with zeolite, on the other hand, its amount

decreases due to mixing and placement at zeolite sites. So, the photocatalytic degradation is also reduced [32-35]. On the other hand, because the zeolite sites are no longer

completely empty and a percentage of them are occupied by  $\text{TiO}_2$ , the surface adsorption of the produced sulfone on the zeolite surface is also reduced [36-39].

*Results of desulfurization experiments with group (c) photo catalysts*

**Table 10:** Comparison of the efficiency of group C photo catalysts in desulfurization

Efficiency	Radiation time	Type of radiation	Amount and concentration of $\text{H}_2\text{O}_2$	mass (gr)	Catalyst type
17.98%		UV-C		0.3	<b>Pcat(10)</b>
23.49%		UV-C	-35% ml0.2	0.5	<b>Pcat(10)</b>
99.45%	4hours	Visible		0.5	<b>Pcat(11)</b>

Ce metal, as a dopant, has a much better performance and higher efficiency in the desulfurization process than Ag metal [40-42].

*Results of desulfurization experiments with group (d) photo catalysts*

**Table 11:** Comparison of the efficiency of group D photo catalysts in desulfurization

Efficiency	Radiation time	Type of radiation	Amount and concentration of $\text{H}_2\text{O}_2$	Mass (gr)	Catalyst type
15.72%				0.05	<b>Pcat (12)</b>
19.61%				0.1	
25.31%				0.3	
36.31%	4hours	UV-C	-35%ml 0.2	0.5	
14.09%				0.7	
20.50%				0.1	<b>Pcat (13)</b>
36.42%				0.3	
34.81%				0.1	<b>Pcat (14)</b>
36.47%				0.3	

**Table 12:** Relationship between  $\text{TiO}_2$  (P25) doped in photo catalyst with DBT reduction percentage

Efficiency	Percentage of $\text{TiO}_2$ (P25) doubled in photo catalyst	Catalyst type
36.31%	54.32%	<b>Pcat(12)</b>
36.42%	27.06%	<b>Pcat(13)</b>
36.47%	136.62%	<b>Pcat(14)</b>

**Table 13:** Comparison of the efficiency of photo catalysts in section (e-I) in desulfurization

Efficiency	Radiation time	Type of radiation	Amount and concentration of $\text{H}_2\text{O}_2$	Mass (gr)	Catalyst type
17.50%		UV-C		0.3	<b>Pcat(15)</b>
21.95%		UV-C		0.5	<b>Pcat(15)</b>
9.63%		UV-C		0.5	<b>Pcat(16)</b>
17.09%	4hours	Visible	-35% ml0.2	0.5	<b>Pcat(16)</b>
0%		Visible		0.5	<b>Pcat(17)</b>
33.52%		Visible		0.5	<b>Pcat(18)</b>
35.65%		Visible		0.5	<b>Pcat(19)</b>
60.69%		Visible		0.5	<b>Pcat(20)</b>
99.40%		Visible		0.5	<b>Pcat(21)</b>

*Percentage of different weights of cerium and silver metals in the active component ZnO, ZnS and CdS*

The photo catalysts synthesized in Section (II) contain doped amounts of different amounts of cerium dopant in the active component of zinc oxide and sulfide, as well as silver doped in cadmium sulfide [43-46].

**Table 14:** Comparison of the efficiency of section (e-II) photo catalysts in desulfurization

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst type
0%		Visible			Pcat(22)
36.15%		Visible			Pcat(23)
0%	4hours	Visible	-35% ml0.2	0.5gr	Pcat(24)
2.84%		Visible			Pcat(25)
7.1%		UV-C			Pcat(25)
14.905		Visible			Pcat(26)

**Table 15:** Comparison of the efficiency of "T" group photo catalysts in desulfurization

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst type
80.51%					Pcat(27)
99.97%	4hours	Visible	-35% ml0.2	0.5gr	Pcat(28)
99.99%					Pcat(29)

*Determining the type of process used in this research to desulfurize*

As a result of the use of zeolite-based photo catalysts in the oxidative desulfurization

process, the question arises as to whether the desulfurization efficiency may have been achieved solely as a result of the adsorption of the DBT compound on the zeolite surface.

**Table 16:** Desulfurization efficiency solely as a result of the adsorption process on zeolite

Efficiency	Radiation time	Type of radiation	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass (gr)	Catalyst type
13.88%	-	4hours	Nanoxelite NaX	-35% ml0.2	-

In addition, when the desulfurization mechanism is supposedly based on the DBT adsorption process on the catalyst particles, the process efficiency is lower in cases where the sample is oxidized than in the non-oxidant condition because the oxidant is sometimes used as the oxidant. In sulfur removal, light radiation (ultraviolet or visible) will be present in the sample, in other words, the desulfurization mechanism is due to the photocatalytic process. Otherwise, when the mechanism of action is

based on surface adsorption, H<sub>2</sub>O<sub>2</sub> is considered as a competitive agent for adsorption on the catalyst, and if the active component of the catalyst is TiO<sub>2</sub>, because TiO<sub>2</sub> is polar and H<sub>2</sub>O<sub>2</sub> is more polar than DBT. So naturally it will be easier to absorb TiO<sub>2</sub> [47-49].

H<sub>2</sub>O<sub>2</sub> is also smaller in size and can sit in more active locations than the catalyst, but the DBT molecule is large and, when placed in a TiO<sub>2</sub> site, also occupies the space of its side sites, thus

reducing its absorption and therefore replenishing it.

### Calculation of dipole moment of di-benzothiophene and di-benzothiophene sulfonate compounds by quantum chemistry theory method

Quantum chemistry calculations were used to calculate the dipole moment of raw material and product. First, the geometric structures of two combinations of d-benzothiophene and d-benzothiophene sulfonate were created and the bipolar moment of both combinations was optimized using the following command line in Gaussian 2009 software.

MP2 /6-31 G\* Opt Symm = (Scf , PG =C<sub>2v</sub> , follow) test

$$\mu_{(DBT)} = 1.1675 \text{ (D)}$$

$$\mu_{(DBT-Sulfone)} = 6.4257 \text{ (D)}$$

As can be seen from the calculations above, the polarity of the dibenzothiophene sulfone compound produced during the photocatalytic degradation process was 5.5 times higher than the polarity of the DBT raw material, which is due to the adsorption process of the dibenzothiophene sulfone compound on the surface of fugasite nano zeolite. NaX is very effective. Thus, when the DBT compound is oxidized to its corresponding sulfone in the photocatalytic process, the degree of polarity is increased so that more adsorption of the zeolite will occur.

**Table 17:** Results of kinetic experiments with photo catalyst (Ni (8%) / TiO<sub>2</sub> / zeolite NaX)

Efficiency	qt (mg/g)	Final concentration (ppm)	Type of radiation	Radiation time (min)	Initial concentration (ppm)	Amount and concentration of H <sub>2</sub> O <sub>2</sub>	Mass	Catalyst type
80.28%	1.6328	20.05		50				
98.57%	2.0048	1.450		100				
99.59%	2.0256	0.41	Visible	150	101.696	-35% ml0.2	0.5gr	<b>Pcat(29)</b>
99.98%	2.0334	0.02		200				
99.995	2.0336	0.01		250				

### 3) Blanchard equation:

$$1 / (q_e - q_t) = 1 / q_e + k_2 t$$

The results of the graphs related to kinetic equations along with their correlation coefficients are presented in table (18).

**Table 18:** Results of diagrams related to kinetic equations

Correlation coefficient (R <sup>2</sup> )	Regression equation	The chart	General formula	Synthetic model
0.967	Y = -0.048x + 1.570	Ln (q <sub>e</sub> - q <sub>t</sub> ) in terms of t	dq <sub>e</sub> /dt = k <sub>1</sub> (q <sub>e</sub> - q <sub>1</sub> )	<b>Lagergren</b>
0.825	Y = 0.294x + 0.540	q <sub>t</sub> in terms of ln t	q <sub>t</sub> = 1/β Ln(αβ) + 1/β Ln(t)	<b>Al-Wawich</b>
0.619	Y = 30.16x - 2480	1 / q <sub>e</sub> q <sub>t</sub> in terms of t	1/q <sub>t</sub> = 1/k <sub>2</sub> q <sub>e</sub> <sup>2</sup> + 1/q <sub>e</sub>	<b>Blanchard</b>

### Conclusion

According to the above tables, it is concluded that the experimental data do not match the quasi-quadratic kinetics on a line and have a small correlation coefficient. The result of the kinetic analysis of the oxidative reaction of the

sulfur compounds of the model shows that the highest correlation coefficient belongs to the Lagergren model, which is the quasi-first-order kinetic model. Following the reaction of quasi-first-order kinetics means that in fact the reaction rate is dependent on the concentration of the two substances, which by considering one



of the concentrations higher than the other, the effect is eliminated in studying the reaction rate. Is a function of the concentration of the DBT compound and changes in H<sub>2</sub>O<sub>2</sub> concentration have no effect on the reaction rate?

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