

## Oxidative Desulfurization of Dibenzothiophene Using Dawson Type Heteropoly Compounds/Tantalum as Catalyst

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### ABSTRACT

Catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  has been synthesized by simple wet impregnation at 30-40 °C under atmospheric conditions using Dawson type polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]$  and tantalum. The catalyst was characterized by FTIR spectrophotometer, XRD, SEM, and  $\text{N}_2$  adsorption desorption methods. FTIR spectrum of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  showed that Dawson type polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]$  and Ta was successfully impregnated which was indicated by vibration spectrum at wavenumber of 900-1100  $\text{cm}^{-1}$  for polyoxometalate and 550  $\text{cm}^{-1}$  for Ta. The surface area of the  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  after impregnation was higher than  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  and its morphology was found to be uniform. The catalytic activity of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  toward desulfurization of dibenzothiophene was three times higher than the original catalyst of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  without impregnation. The catalytic regeneration test of catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  showed that the catalytic activity for first regeneration of catalyst has similar catalytic activity with the fresh catalyst without loss of catalytic activity indicated by almost similar percent conversion.

**Keywords:** dibenzothiophene; Dawson type; heteropoly; tantalum; desulfurization

### ABSTRAK

Katalis  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  telah disintesis dengan impregnasi basah sederhana pada temperatur 30-40 °C pada kondisi atmosfer menggunakan senyawa polioksometalat Tipe Dawson  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]$  dan tantalum. Katalis dikarakterisasi dengan FTIR, XRD, SEM, dan metode adsorpsi desorpsi  $\text{N}_2$ . FTIR spectrum  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  menunjukkan bahwa senyawa polioksometalat tipe Dawson  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]$  dan Ta telah berhasil diimpregnasi yang ditunjukkan munculnya bilangan gelombang 900-1100  $\text{cm}^{-1}$  untuk polioksometalat dan 550  $\text{cm}^{-1}$  untuk Ta. Hasil karakterisasi diperoleh luas permukaan katalis  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  setelah impregnasi meningkat dibandingkan dengan  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  dengan bentuk morfologi seragam. Aktivitas katalitik  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  terhadap reaksi desulfurisasi dibenzothiophen menunjukkan tiga kali lebih tinggi dari pada katalis  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  tanpa impregnasi. Pengujian penggunaan ulang katalis  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  pada penggunaan ulang pertama menunjukkan hasil yang sama dengan katalis awal tanpa kehilangan aktivitas katalitik yang ditunjukkan dengan kemiripan persen konversi.

**Kata Kunci:** dibenzothiophen; Dawson type; heteropoli; tantalum; desulfurisasi

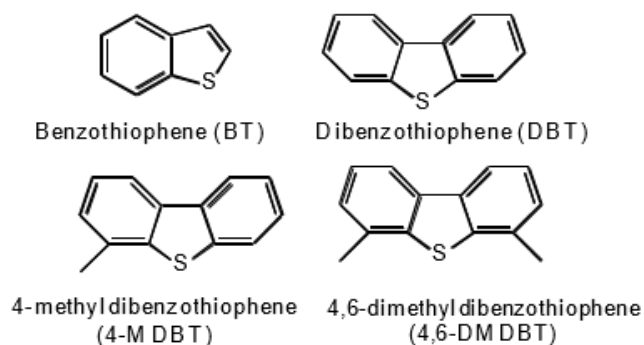
### INTRODUCTION

Polyoxometalates have been intensively used as catalyst in organic synthesis and transformations including acid base and oxidation-reduction reactions. These properties are due to various kind of oxidation states, variation of structures including flexibility to change addenda or heteroatoms, applicable in homogeneous or heterogeneous reaction systems, and its ability to support or impregnated with many kinds of

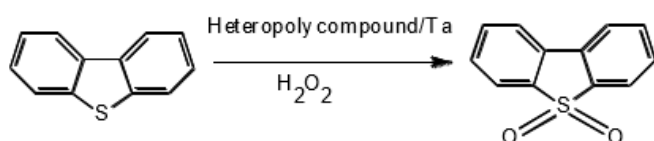
materials [1]. Supporting polyoxometalate on other solids such as metal oxide improves not only heterogeneous properties but also catalytic performance for organic reactions and transformations. Polyoxometalate supported metal oxide was prepared because strong interaction of active sites with supporting material which resulted into high surface area and catalytic properties [2].

The organic reaction and transformation have attracted a lot of attention related to the environmental

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**Fig 1.** Structures of common organosulfur in fuel



**Fig 2.** Oxidative desulfurization of dibenzothiophene to the corresponding sulfone

global issue such as pollution of sulfur from transportation fuel based fossil [3]. Sulfur in transportation fuel came from organic sulfur which is called organosulfur such as benzothiophene (BT), dibenzothiophene (DBT), 4-methyl dibenzothiophene (4-MDBT), and 4,6-dimethyl dibenzothiophene (4,6-DMDBT) as shown in Fig. 1. Desulfurization of these compounds using hydrodesulfurization (HDS) has not been effective [4]. The HDS is very effective for organosulfur with less steric hindrance and no refractory index. On the other hand, HDS is operated industrially at high temperature and pressure [5].

Efficient desulfurization that was promising to carry out is oxidative desulfurization (ODS). In ODS process, the refractory organosulfur such as BT, DBT, 4-MDBT, and 4,6-DMDBT were successfully converted into the corresponding sulfone at mild conditions using appropriate catalyst [6]. Green oxidant such as hydrogen peroxide or molecular oxygen is usually used in the reaction due to formation of water as byproduct. Sulfone from the reaction mixture was easily removed by physical techniques such as adsorption, extraction, filtration, or distillation. The successful key for ODS is suitable catalyst for reaction to form sulfone [7]. In continuation of our group research on synthesis of supported polyoxometalate and its application is reported here, the supported tantalum into Dawson type polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  via wet impregnation at 30-40 °C [8]. The chemical characterization was carried out using FTIR spectroscopy, X-Ray powder analysis, morphology analysis by SEM, and porosity through  $\text{N}_2$  sorption measurement. The catalytic performance of

catalyst was tested on ODS of BT at mild conditions. The reaction scheme of this process was illustrated in Fig. 2.

## EXPERIMENTAL SECTION

### Materials

All chemical reagents were used directly after purchase such as dibenzothiophene (Sigma-Aldrich), sodium tungsten (Merck), orthophosphoric acid (Merck), ammonium chloride (Merck), ethanol (Merck), hydrogen peroxide (Merck), and tantalum(V) chloride (Sigma-Aldrich).

### Instrumentation

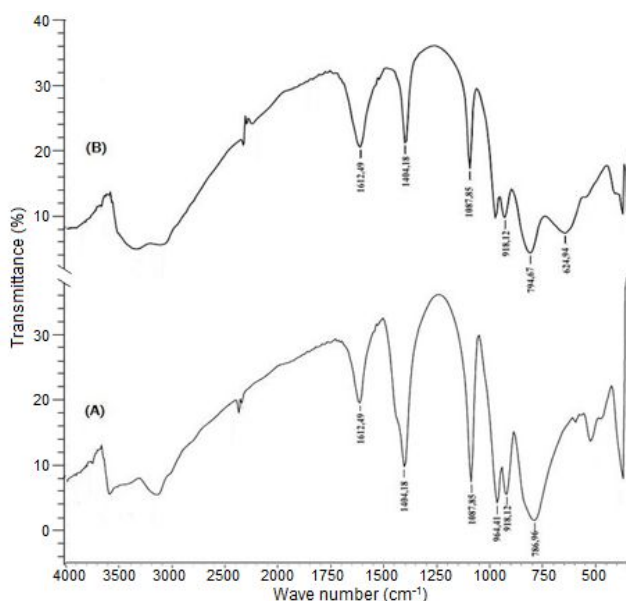
Measurement of IR spectrum was performed using FTIR Shimadzu Prestige-21 spectrophotometer with KBr disc and the data was collected at 300-4000  $\text{cm}^{-1}$ . X-Ray analysis was conducted using Shimadzu Lab X Type 6000 diffractometer and the data was acquired over  $2\theta$  0-90 deg. Textural properties were performed using JEOL JED-2300 SEM analysis station. Nitrogen adsorption desorption was carried out using AutosorbIQ automated gas sorption analyzer and surface area measurement was calculated using BET analysis.

### Procedure

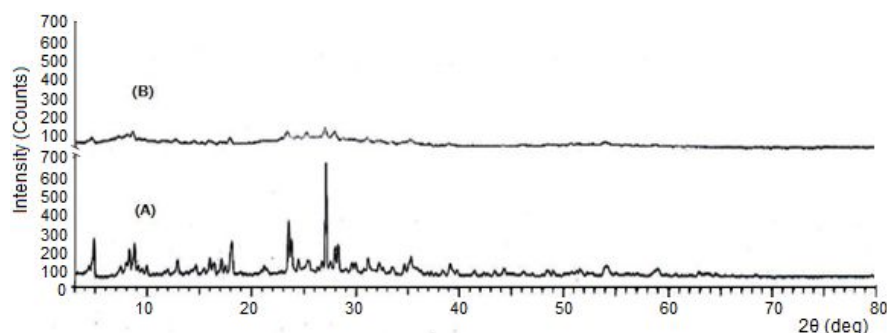
#### Synthesis of polyoxometalate

##### $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$ and characterization

Polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  was synthesized according to Contant et al. [9] as follows. In a 2 L beaker glass a sample of 250 g sodium tungsten was dissolved in 500 mL of water and 210 mL of orthophosphoric acid was added. The solution was refluxed for 4 h. After cooling, 100 g of ammonium chloride was added, and the solution was stirred for 10 min. The pale yellow salt was removed by filtration, dissolved in 600 mL of water and precipitated again with 100 g of ammonium chloride. After stirring for 10 min, the solution was filtered. The precipitate was dissolved in 250 mL of warm water to obtain clear solution. The solution was kept for 5 days to produce crystals of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$ . Recrystallization was performed using warm water at pH 2 using addition of hydrochloric acid to obtain white crystals of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$ . Dawson type polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  was characterized using FTIR spectroscopy, X-Ray analysis, and SEM measurement.



**Fig 3.** FTIR spectrum of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  (A) and  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  (B)



**Fig 4.** XRD powder pattern of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  (A) and  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  (B)

#### **Synthesis of $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$ and characterization**

Catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  was prepared according to the procedure from Lesbani et al. [8] which modified from Xu et al. [10]. In 50 mL of beaker glass, 0.5 g of polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  was dissolved in 5 mL of ethanol with stirring and 2.5 mL of water was added (solution A). In separate beaker glass, 0.6 g of tantalum chloride was dissolved in 5 mL of ethanol (solution B). The solution A was added into the solution B with slow stirring for 1 h. Continuous stirring was maintained under 30-40 °C for 24 h to form sol homogeneous materials. The sol materials were heated at 110 °C for 3 h to obtain  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$ . The resulting  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  was then characterized using FTIR, X-Ray, and SEM analyses.

#### **The catalytic oxidative desulfurization of dibenzothiophene by $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$ catalyst**

Catalytic oxidative desulfurization of dibenzothiophene was carried out in 100 mL of Schlenk flask equipped with a magnetic stirrer and hot plate;

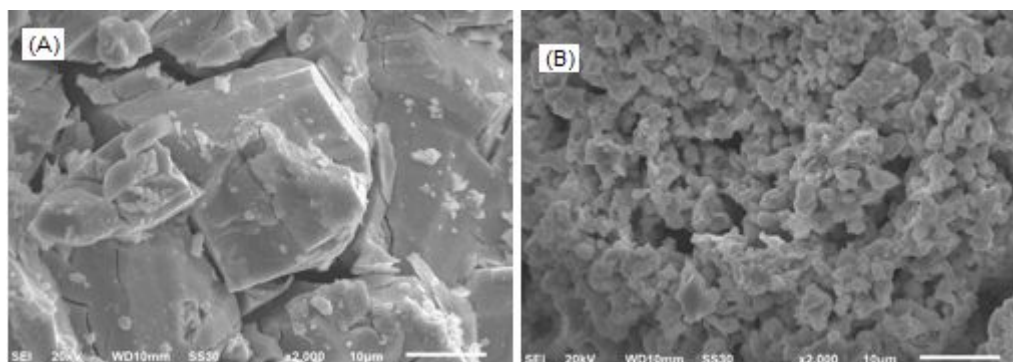
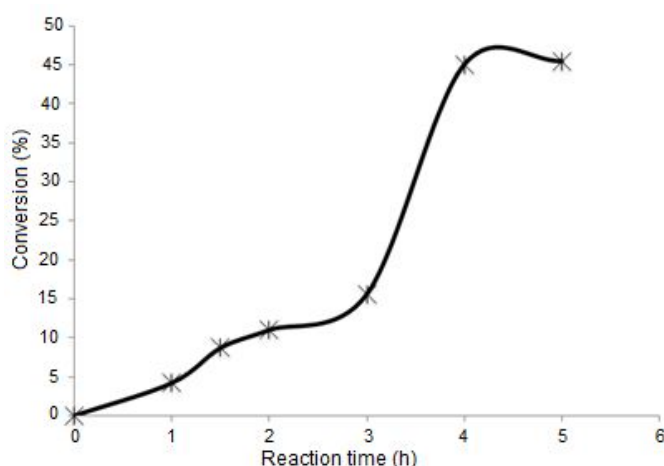
0.1 g of dibenzothiophene was mixed with 5 mL of hexane, 0.5 mL hydrogen peroxide, and 0.1 g of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  catalyst. Reaction was stirred at 70 °C for 2 h. The formation of sulfone was monitored using GC equipped with RTX-1 capillary column. The initial reaction was also checked using GC. Conversion of dibenzothiophene to corresponding sulfone compound was calculated according to formulation from Lesbani et al. [11]. Desulfurization of dibenzothiophene was further investigated through variation of reaction time, temperature reaction, and weight of catalyst.

## **RESULT AND DISCUSSION**

The FTIR spectra of polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  and  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  are presented in Fig. 3. The FTIR spectra of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot n\text{H}_2\text{O}$  showed bands at 786  $\text{cm}^{-1}$  ( $\nu\text{W-O}_e\text{-W}$ ), 918  $\text{cm}^{-1}$  ( $\nu\text{W-O}_e\text{-W}$ ), 964  $\text{cm}^{-1}$  ( $\nu\text{W=O}$ ), 1087  $\text{cm}^{-1}$  ( $\nu\text{P-O}$ ), and 1404  $\text{cm}^{-1}$  ( $\nu\text{N-H}$ ). Another vibration at

**Table 1.** Surface area properties of heteropoly compound/Ta catalyst

Compound	Surface area ( $S_{\text{BET}}$ , $\text{m}^2/\text{g}$ )	Pore volume ( $V_{\text{BJH}}$ , $\text{cm}^3/\text{g}$ )	Pore diameter ( $\varnothing_{\text{BJH}}$ , nm)
$(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot\text{nH}_2\text{O}$	2.05	0.003	3.008
$(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$	75.42	0.087	2.424

**Fig 5.** SEM morphology of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot\text{nH}_2\text{O}$  (A) and  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  (B)**Fig 6.** Effect of reaction time to oxidative desulfurization using heteropoly compound/Ta catalyst

$1612\text{ cm}^{-1}$  also indicated the existing of N-H from  $\text{NH}_4$  [12]. The supported polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot\text{nH}_2\text{O}$  on tantalum to form  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  resulted the FTIR spectrum similar with the original polyoxometalate. The process of impregnation was identified using XRD powder analysis as shown in Fig. 4.

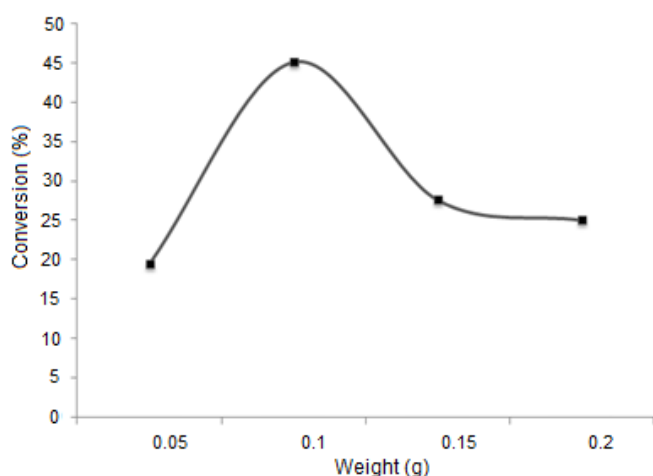
The XRD diffraction pattern of polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot\text{nH}_2\text{O}$  indicated high crystallinity of these Dawson type polyoxometalate. The crystallinity appeared at diffraction less than 10 deg which possess unique and specific diffractions for polyoxometalates [13]. The diffraction of tantalum(V) chloride as shown in Fig. 4B was for amorphous material with less diffraction peak at wide diffraction angle. The combination of crystalline material with amorphous material in this research resulted into catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$

with high crystallinity (Fig. 4A). Several diffraction peaks were identified in relation to the formation of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  at 20-30 deg.

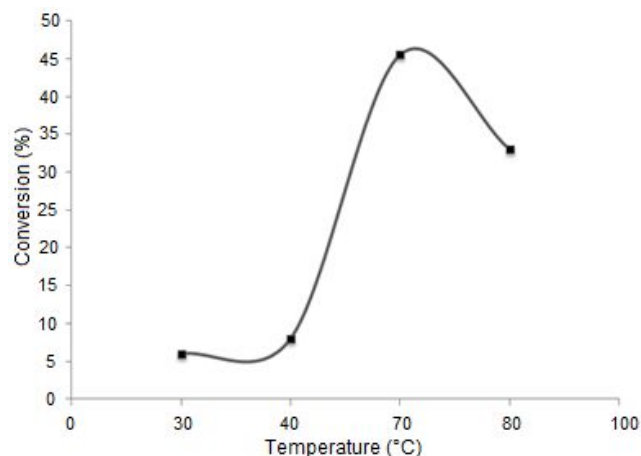
Morphology of catalyst before and after impregnation was also observed using SEM analysis. The surface photo analysis of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot\text{nH}_2\text{O}$  and  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot\text{nH}_2\text{O}$  are presented in Fig. 5. The morphology of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]$  after supported with tantalum indicated uniform properties with small size particles. By supporting tantalum, the regular shape form indicated the increase surface area properties as shown in Table 1. The surface area properties of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  showed sharp increase after  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot\text{nH}_2\text{O}$  impregnated with tantalum compared to the original surface area of polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot\text{nH}_2\text{O}$ . High surface area is commonly found in all structure and types of polyoxometalates which resulted many polyoxometalates to be suitable in homogeneous catalytic rather than heterogeneous catalytic system [14].

The catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  was used as heterogeneous catalyst for desulfurization of dibenzothiophene under 70 °C. The catalytic activity was monitored to evaluate the catalytic activity of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$ . Conversion of benzothiophene to corresponding sulfone at initial reaction was relatively slow, which then increase gradually corresponding to time as shown in Fig. 6. Conversion of polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]\cdot\text{nH}_2\text{O}$  as catalyst was very low as shown in Table 2 and relatively moderate after impregnated with tantalum.

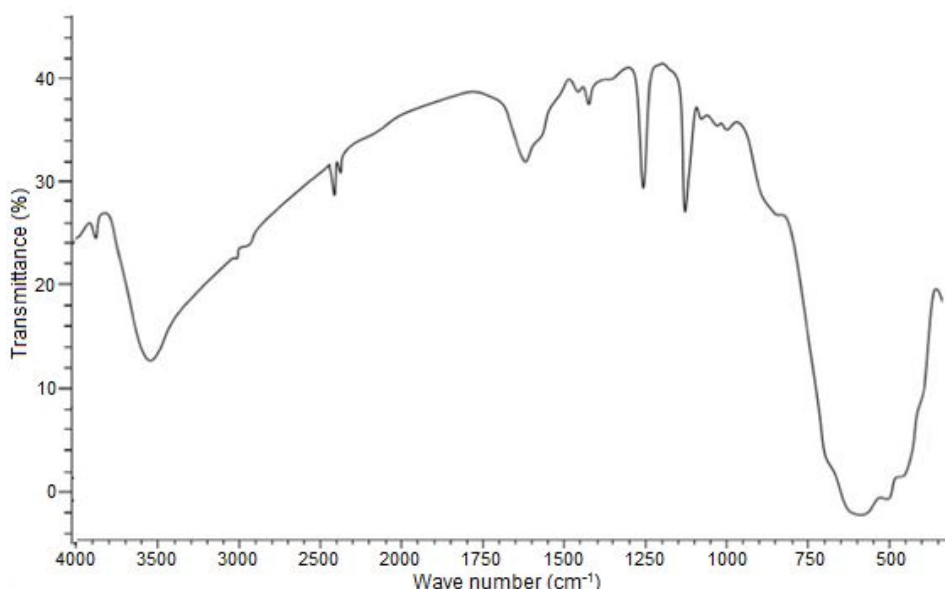
The rate of reaction was relatively constant after 5 h. The conversion of dibenzothiophene at 5 h was up to 45% and it was similar with reaction at 4 h, which



**Fig 7.** Effect of catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  weight for oxidative desulfurization of dibenzothiophene



**Fig 8.** Effect of temperature on conversion of oxidative desulfurization of dibenzothiophene by catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$



**Fig 9.** FTIR spectrum of catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  after used in oxidative desulfurization of dibenzothiophene

**Table 2.** Catalytic activity of heteropoly compound/Ta catalyst in oxidative desulfurization of dibenzothiophene

Catalyst	Conversion (%)
$(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}] \cdot n\text{H}_2\text{O}$	15.0
$(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$ (Fresh catalyst)	45.5
$(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$ (1 <sup>st</sup> regeneration)	45.0

resulted into the reaction to be stopped at 5 h. The possible reason to explain this result is the active site of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  was saturated with dibenzothiophene which caused slower reaction process in this catalytic system. To evaluate all catalytic reaction system, several factors in the reaction flask have been investigated, such as effect of catalyst weight,

temperature, and reuse of catalyst. The effect of catalyst weight was shown in Fig. 7.

The weight of catalyst 0.1 g is less than 1/10 mol or mmol dibenzothiophene. According to general catalytic theories that the weight used in this research is appropriate to the catalyst weight requirement as a catalyst. In this research, we found that 0.1 g is the optimum catalyst weight for conversion of dibenzothiophene to sulfone. The weight of catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  more than 0.1 g was found to be ineffective for conversion of dibenzothiophene.

The effect of temperature for conversion of dibenzothiophene were presented in Fig. 8 and at temperature of 80 °C was able to produce an optimum of percent conversion of dibenzothiophene over

$(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  catalyst up to 45%. Temperature at near room temperature condition was not suitable for the oxidation of dibenzothiophene. Lesbani et al. [14] found that the optimum temperature condition for desulfurization of benzothiophene was at 40 °C, which was a relatively lower temperature condition than it was operated in this research.

The performance of fresh catalyst and after regeneration process was also deeply investigated. The catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  after first use was washed with demin water and dried under 110 °C and used for the second reaction of desulfurization. The catalytic reaction activity of the fresh catalyst compared to catalyst after regeneration produce almost the same amount of percent conversion. The data of catalytic regeneration of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  is shown in Table 2.

The data in Table 2 shows similar catalytic activity of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  was found in this reaction system. There was almost no difference in catalytic activity for desulfurization of dibenzothiophene under catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  for fresh and first regeneration of catalyst. The FTIR spectra of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  before it was used as catalyst (Fig. 3B) and after catalytic reaction were equivalent as shown in Fig. 9. All specific vibration of polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}] \cdot n\text{H}_2\text{O}$  were also found in the catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  FTIR spectra after usage in the reaction of oxidative desulfurization of dibenzothiophene which indicate the stability of the catalyst [15].

## CONCLUSION

Polyoxometalate  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}] \cdot n\text{H}_2\text{O}$  was successfully impregnated with tantalum to form  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  with increase in surface area and regular morphology size. The catalytic activity of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  to aid oxidative desulfurization of DBT was three times higher than  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}] \cdot n\text{H}_2\text{O}$  before being impregnated with tantalum. The catalytic properties of  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  at first regeneration produce almost similar percent conversion to the fresh catalyst, which indicate that the catalyst  $(\text{NH}_4)_6[\beta\text{-P}_2\text{W}_{18}\text{O}_{62}]/\text{Ta}$  possess stable catalytic activity at reaction temperature of 80 °C.

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