

Synthesis of Ce-Mesoporous Silica Catalyst and Its Lifetime Determination for the Hydrocracking of Waste Lubricant

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ABSTRACT

The synthesis of Ce/mesoporous silica (Ce/MS) and its lifetime determination for the hydrocracking of waste lubricant has been carried out. The MS was synthesized using tetraethyl orthosilicate (TEOS) and gelatin extracted from bovine bone as a template. Cerium was impregnated onto the MS by wet impregnation method using $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$. The MS and Ce/MS were then characterized by means of acidity using ammonia base vapor adsorption, Fourier Transform Spectrophotometer (FTIR), Transmission Electron Microscope (TEM), Scanning Electron Microscope-Energy Dispersive X-ray Spectrometer (SEM-EDX), and surface area analyzer (SAA) based on the BET and BJH equation. The Ce/MS catalyst was tested in hydrocracking of waste lubricant in three runs. Lifetime of Ce/MS catalyst was determined using a linear regression of the liquid product yields vs hydrocracking time. The Ce/MS catalyst showed an acidity of 2.79 mmol/g, BJH desorption pore diameter of 3.84 nm, BET surface area of 246.55 m^2/g , and total pore volume of 0.44 cm^3/g . The yield of liquid product obtained from hydrocracking of waste lubricant using the Ce/MS catalyst for the first, second, and third runs was 21.42, 17.23 and 10.54 wt.%, respectively for 2.5 h per each run. Lifetime of Ce/MS catalyst in hydrocracking of waste lubricant was 12.54 h.

Keywords: hydrocracking; waste lubricant; mesoporous silica; catalyst lifetime

ABSTRAK

Telah dilakukan sintesis Ce-silika mesopori (Ce-MS) dan penentuan umurnya untuk hidrorengkah pelumas bekas. Silika mesopori disintesis menggunakan tetraetil ortosilan (TEOS) sebagai prekursor silika dan gelatin yang diekstraksi dari tulang sapi sebagai pencetak. Katalis Ce-MS selanjutnya dianalisis menggunakan FT-IR, TEM, SEM-EDS, dan GSA (Gas Sorption Analyzer). Hasil penelitian menunjukkan bahwa silika mesopori berhasil disintesis dengan diameter pori 3,43 nm, luas permukaan spesifik 286,45 m^2/g , dan volume pori total 0,44 cm^3/g . Katalis Ce-MS telah disintesis dengan diameter pori 3,83 nm, luas permukaan spesifik 260,99 m^2/g , dan volume pori total 0,46 cc/g . Fraksi cair yang diproduksi dari hidrorengkah pelumas bekas berturut-turut sebesar 21,42, 17,23 and 10,54% untuk langkah pertama, kedua, dan ketiga hidrorengkah, di mana setiap langkah membutuhkan waktu selama 2,5 jam. Umur katalis Ce-MS ditentukan menggunakan persamaan regresi linier dari persen produk cair terhadap waktu hidrorengkah, dihasilkan 12,53 jam.

Kata Kunci: hidrorengkah; pelumas bekas; silika mesopore; umur katalis

INTRODUCTION

Waste treatment is the most important concern of modern society to protect the environment. In addition, waste treatment is also an alternative way to overcome other problems faced by modern society. One of the important problems facing modern society is the reduction of fossil fuel sources. Various studies on alternative fuel sources have been conducted [1-2]. One of them is the utilization of waste lubricant as an alternative source of fuel [3-4]. Waste lubricant contains many carbon atoms that can be converted into fuel with special treatment. Waste lubricant contains long chain hydrocarbon compounds ($>\text{C}_{30}$). Therefore, the

hydrocracking process is indispensable in the conversion of waste lubricant into fuel.

Hydrocracking is one of the most important processes in petroleum processing. In the hydrocracking process, the hydrocarbon compound breaks down into smaller-chain hydrocarbon compounds [5-6]. This hydrocracking process requires a catalyst in order to produce as much product as possible in a short time. Transition metal plays an important role in providing acid sites to initiate the hydrocracking reactions [7-8]. However, the use of transition metals directly is not effective because agglomeration and sintering may occur during hydrocracking process. This can be solved by loading

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the transition metal into a support material that has large surface area.

Mesoporous material such as mesoporous silica is an interesting field for being studied as a support material. This mesoporous material has high thermal stability, large surface area, optimum acidity, and high porosity [9-12]. Synthesis of mesoporous silica requires a source of silica and a template. The most commonly used silica source is tetraethyl orthosilicate (TEOS) or sodium silicate, while for the template is a cationic organic compound such as cetyl trimethylammonium bromide (CTAB) [4,10]. However, the use of cationic template for synthesis of mesoporous silica is expensive. As an alternative, nonionic template such as gelatin can be used for synthesis of mesoporous silica [13-14].

In the present work, the authors undertake to synthesize a mesoporous silica (MS) using bovine bone gelatin as a template. Cerium salt solution was loaded onto the MS by wet impregnation method and it was reduced to produce Ce/MS catalyst. The catalysts activity and selectivity in hydrocracking of waste lubricant was evaluated. Lifetime of Ce/MS catalyst was also determined.

EXPERIMENTAL SECTION

Materials

TEOS (99%, p.a), NaOH (p.a), HCl (37%, p.a), CH₃COOH (p.a), NH₄OH (p.a), KI, KIO₃ (p.a), Ce(NO₃)₃·6H₂O (p.a) was purchased from Merck (Germany). The bovine bone was obtained from a local market in Yogyakarta, Indonesia.

Instrumentation

The functional groups of all sample were determined using Fourier Transform Infrared Spectroscopy (FTIR, Shimadzu Prestige-21) equipped with data station in the range of 400–400 cm⁻¹ with a KBr disc technique. Surface area analyzer (SAA, Quantachrome NovaWin Series) was used to determine the surface parameters (surface area, pore volume, and pore diameter) of the MS and Ce/MS. The determination was based on physical adsorption of N₂ gas at batch temperature of 77.3 K. The MS and Ce/MS were degassed at 300 °C for 3 h. The morphology of the MS was taken using a scanning electron microscope (SEM, JEOL JSM-06510LV) equipped with energy dispersive X-ray spectrometer (EDX, JEOL JED-2300) to determine its metal content. Imaging was carried out at 30 kV accelerating voltage using backscattered electron imaging technique. The pore image was taken using transmission electron microscope (TEM, JEOL JEM-1400) at 120 kV accelerating voltage. The liquid

produced by hydrocracking of waste lubricant was analyzed using gas chromatography-mass spectrometry (GC-MS, Shimadzu QP2010S) with a column length of 30 m, diameter of 0.25 mm, thickness of 0.25 μm, temperature of 60–310 °C, Helium gas as carrier gas, and acceleration voltage of 70 Ev.

Procedure

Gelatin extraction

The extraction of gelatin from bovine bone was carried out according to the previous work [13]. The bovine bone was pretreated using 4% CH₃COOH, 1M NaOH, and 1 M HCl. The bovine bone was hydrolyzed using aquadest. The gelatin was analyzed using SDS-PAGE and FTIR.

Synthesis of mesoporous silica

The 5 g TEOS was mixed with 5 mL distilled water and added with 0,1 M HCl solution dropwise until pH 2–2.5 and stirred at 35 °C for 1.5 h. The bovine bone gelatin (0.5 g) was dissolved in 40 mL distilled water and stirred at 35 °C for 3 h. The gelatin solution was added with the TEOS solution dropwise and stirred for 24 h. The formed gel was heated in an autoclave at 100 °C for 24 h. The solid was filtered and calcined at 450 °C for 5 h to produce a mesoporous silica (MS). The MS was then analyzed by FTIR, TEM, and GSA as well as acidity determination.

Acidity determination

The acidity of the MS and Ce/MS catalyst were determined by flowing the ammonia vapor into the sample at vacuum condition for 24 h at room temperature. The acidity value of the MS and Ce/MS catalyst were calculated using the following equation:

$$\text{Acidity Value} = \frac{\text{weight after adsorption} - \text{weight before adsorption}}{\text{weight before adsorption} \times M_r \text{NH}_3}$$

Preparation of Ce/MS catalyst

The Ce metal was supported on the MS by sequential wetness impregnation technique. After impregnation, the catalyst was dried in an oven at 70 °C for 24 h. Finally, the catalyst was reduced at 450 °C for 3 h under H₂ gas stream (flow rate of 15 cc/min). The Ce/MS catalyst was characterized by SEM-EDS, TEM, GSA and acidity determination using ammonia base vapor adsorption.

Catalyst activity test

The waste lubricant was filtered and centrifuged. The pretreated waste lubricant was hydrocracked using the Ce/MS catalyst at 450 °C for 2.5 h under H₂ gas stream (flow rate of 20 cc/min). The waste lubricant/catalyst weight ratio was 100. The

hydrocracking process was repeated 3 times using the same catalyst. Liquid product obtained from the hydrocracking was analyzed using GC-MS.

RESULT AND DISCUSSION

Extraction of Gelatin

Fig. 1(a) present FTIR spectra of gelatin extracted from bovine bone. Generally, FTIR spectra of gelatin has five typical absorption peaks, called amide A, amide B, amide I, amide II, and amide III [15-16]. The FTIR spectra of gelatin extracted in this work show the five typical absorption peaks. Absorption peaks at 3410 cm^{-1} corresponds to N-H and O-H stretching vibration (amide A). Absorption peak at 2939 cm^{-1} refers to CH_2 asymmetric stretching vibration (amide B). Absorption peak at 1651 cm^{-1} corresponds to C=O stretching vibration of the amide group coupled with in-plane N-H bending vibration (amide I). Absorption peak at 1543 cm^{-1} refers to in-plane N-H bending and C-N stretching vibration (amide II). Finally, absorption peak at 1242 cm^{-1} refers to combination peaks between C-N stretching vibration and N-H deformation from amide linkages (amide III). These absorption peaks indicate that the gelatin was successfully extracted from bovine bone.

Molecular weight range of gelatin was determined by SDS-PAGE. Gelatin extracted from bovine bone has a molecular weight range of 83–182 kDa. This result show that bovine bone gelatin consists of a mixture of polypeptide from type I collagen with more α -chains (60–120 kDa). The gelatin α -chains consist of polar and non-polar areas. The non-polar areas are made from the tripeptide Gly-Pro-R, where R is a non-polar amino acid, predominantly hydroxyproline. The non-polar areas are interspersed with polar areas, which are relatively deficient in both proline and hydroxyproline. The presence and distribution of the charged, polar and non-polar amino acids provides gelatin with unique properties. The gelatin is easily dissolved in water at the right conditions and forms colloidal solutions [13].

Characterization of MS

Fig. 1(b) and 1(c) showed the FTIR spectra of mesoporous silica before and after calcination. The typical absorption peaks of mesoporous silica was shown at 1096 , 795 , and 471 cm^{-1} that represents the symmetric stretching vibration of Si-O-Si, symmetric bending vibration of Si-O-Si, and bending vibration of Si-O-Si, respectively [4,13-14]. Absorption peak at 3433 and 1636 cm^{-1} represents stretching and bending vibration of Si-OH, respectively. The absorption peak at 2932 and 1543 cm^{-1} refers to amide B and amide II from bovine bone gelatin. After calcination, the absorption peak for amide II was disappeared. The absence of amide II absorption peak indicates that the gelatin was successfully removed from the MS framework. However, the absorption peak at 2924 cm^{-1} still existed after calcination process and it was probably due to the contamination of equipment.

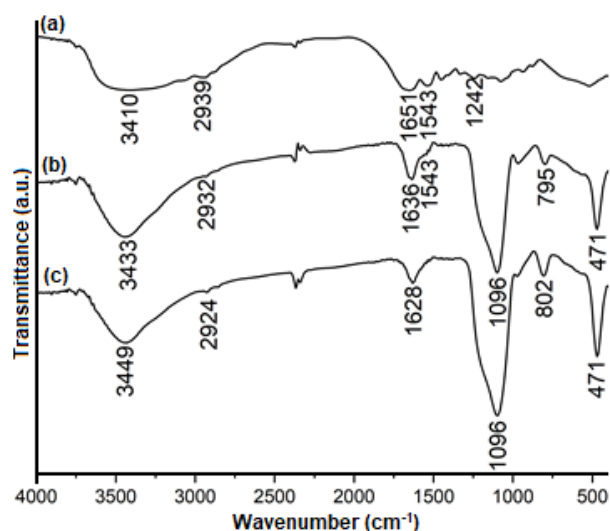


Fig 1. FTIR spectra of (a) bovine bone gelatin, (b) MS, and (c) calcined MS

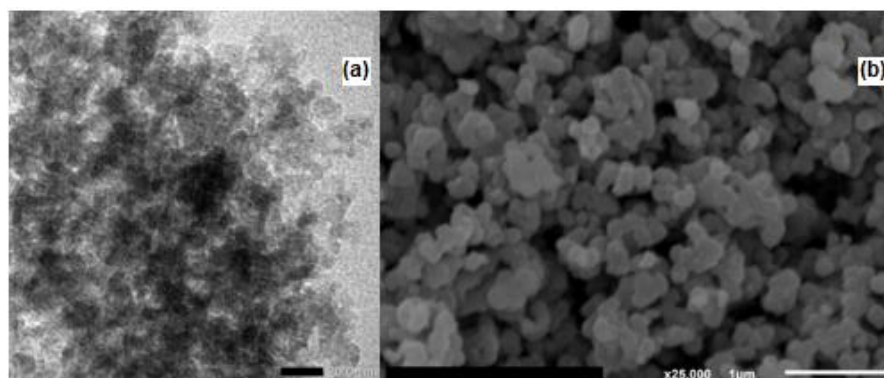


Fig 2. (a) TEM and (b) SEM micrograph of the MS

Characterization of Ce/MS Catalyst

Morphological of MS was shown through TEM and SEM micrographs. The TEM micrograph was shown in Fig. 2(a). The TEM micrograph show the formation of pore on the MS. The pore size is not uniform. It is because the gelatin used as a template in synthesis of MS has wide molecular weight range. Fig. 2(b) presents SEM micrograph of the MS. The 3D morphology of the MS was rounded and low arrangement. It was caused by the irregularity of micelles size from gelatin as a template in synthesis of MS [13]. The impregnation of Ce in MS was proven using EDS. Amount of Ce metal impregnated into the MS was about 1.28%.

Fig. 3(a) and 3(b) present adsorption-desorption of N₂ gas on the MS and the Ce/MS catalyst, respectively. Adsorption-desorption isotherm of N₂ gas on the MS and

the Ce/MS catalyst are a type IV isotherm that indicate the formation of mesoporous [17]. The adsorption-desorption isotherm of N₂ gas on the MS and the Ce/MS catalyst has the H3 hysteresis loop. The H3 hysteresis loop indicate the occurrence of capillary condensation on mesoporous and formation of slit-shaped pores. Fig. 4(a) and 4(b) present pore size distribution of the MS and the Ce/MS catalyst, respectively. Both of the MS and the Ce/MS catalyst have a wide pore size distribution. It is because the molecular weight range of the gelatin that has been explained previously.

Table 1 shows BJH desorption pore diameter, total pore volume, and BET surface area of the MS and Ce/MS catalyst. The pore diameter, pore volume, and surface area of the MS decrease after impregnation of Ce metal. This makes sense because the Ce metal load

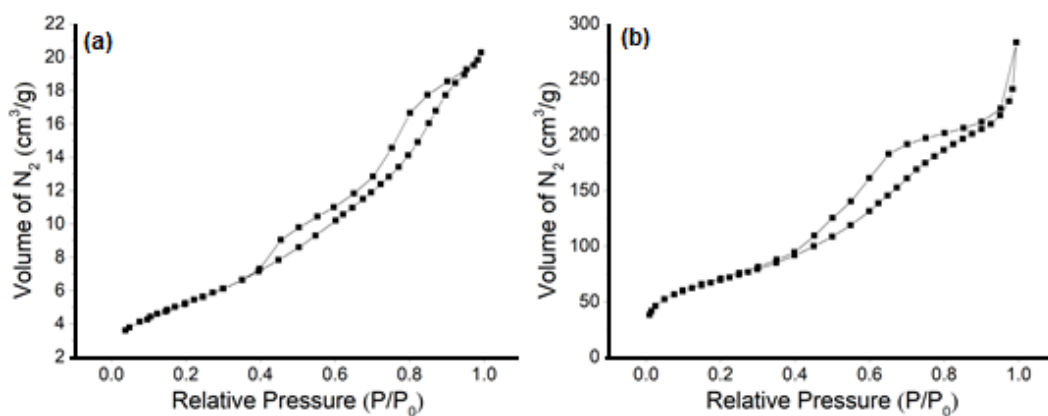


Fig 3. Sorption isotherm of (a) MS and (b) Ce/MS catalyst

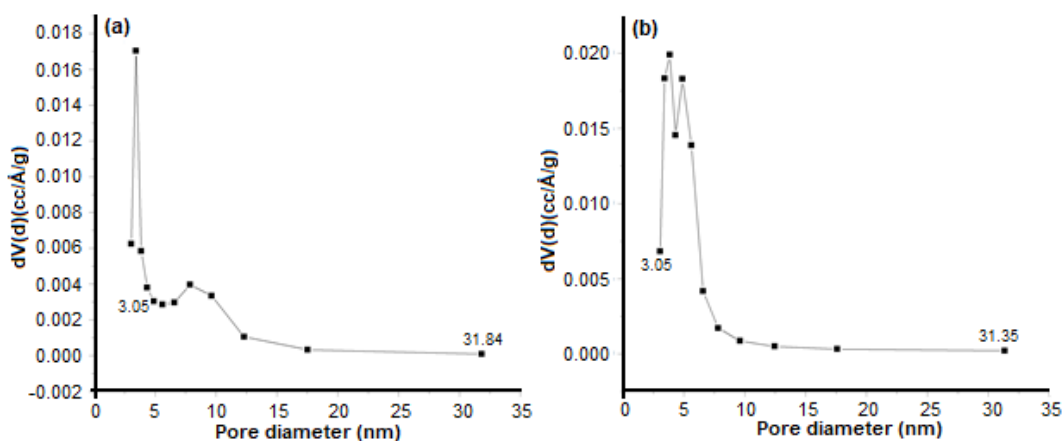


Fig 4. Pore size distribution of (a) MS and (b) Ce/MS catalyst

Table 1. Properties of the MS and Ce/MS

Sample	Acidity value (mmol/g)	BJH desorption pore diameter (nm)	Total pore volume (cm ³ /g)	BET surface area (m ² /g)
MS	1.89	3.42	0.46	279.06
Ce/MS	2.79	3.84	0.44	246.55

into the MS will cover half of the pore wall. The pore diameter data confirms that the MS and Ce/MS catalyst have mesopore according to IUPAC classification (pore diameter of 2–50 nm for mesopore). Acidity of the MS and Ce/MS catalyst was determined gravimetrically by adsorption of ammonia vapor at vacuum condition. The result was shown in Table 1. After impregnation of Ce metal, the acidity of MS increased from 1.89 mmol/g to 2.79 mmol/g. The Ce metal can increase Lewis acid site of the MS by providing empty d orbital. The empty d orbital can accept free electron pair from ammonia vapor [3].

Catalyst Activity Test

Product distribution of hydrocracking of waste lubricant using the Ce/MS catalyst is presented in Fig. 5. Thermal hydrocracking of waste lubricant was carried out as a comparison for evaluating the Ce/MS catalyst activity. On the first run, hydrocracking of waste lubricant using the Ce/MS catalyst produced more liquid product than the thermal hydrocracking. The increase of amount of liquid product in hydrocracking of waste lubricant

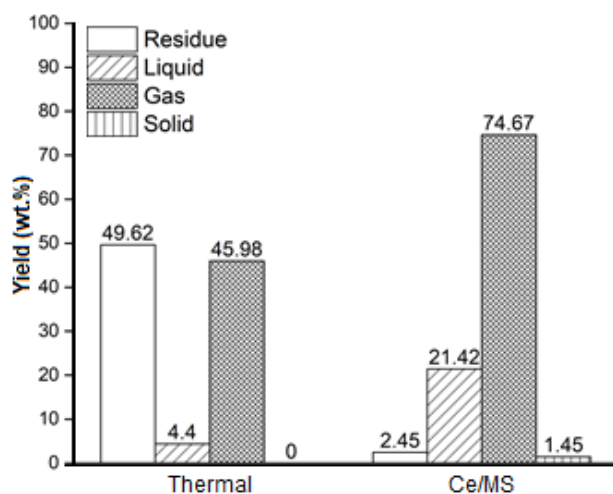


Fig 5. Product distribution of hydrocracking of waste lubricant

using the catalysts can be explained due to its acidic properties. The catalyst synthesized in this work has Lewis acid sites from the Ce metal and Brønsted acid sites from the MS. These acid sites were able to hydrocrack the waste lubricant via carbonium intermediate. Initiation of hydrocracking reaction can occur by proton addition (Brønsted acid) or removing hydrogen anion by Lewis acid that can convert olefinic linkage into an on-chain carbonium. The carbonium chain might be broken up and form short chain hydrocarbon compounds [18-20]. These short chain hydrocarbon compounds can be gas product (C₁-C₄), gasoline (C₅-C₁₂), diesel (C₁₃-C₁₇) or others (>C₁₇).

In Fig. 6, it can be observed that hydrocracking of waste lubricant using the catalysts produced higher amount of fuel product (gasoline and diesel) than thermal hydrocracking. It is caused by acidic properties of Ce/MS catalyst which has been described previously. The acidic properties of Ce/MS catalyst makes the hydrocracking of waste lubricant produced more liquid product. It leads to the fact that more liquid product obtained from hydrocracking of waste lubricant means more fuel products (gasoline and diesel). The

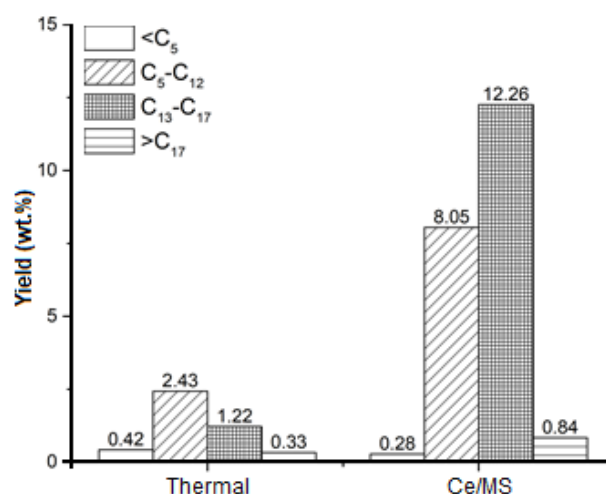


Fig 6. Catalyst selectivity towards gasoline and diesel product

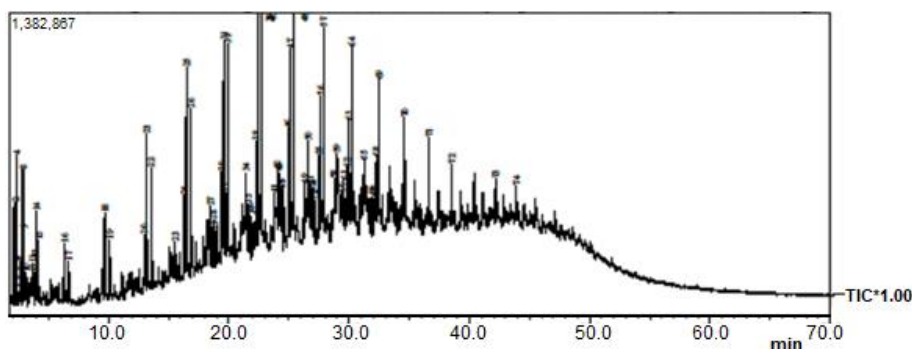
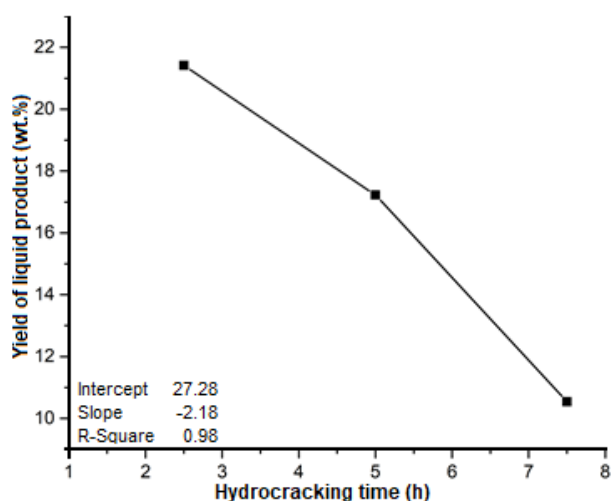


Fig 7. Chromatogram of liquid product from the first run of hydrocracking of waste lubricant

Table 2. Product distribution of hydrocracking of waste lubricant using the Ce/MS catalyst

Run	Conversion (wt.%)			
	Liquid	Gas	Solid	Residue
First	21.42	74.67	1.46	2.45
Second	17.23	75.90	1.38	5.49
Third	10.54	86.22	1.33	1.91

**Fig 8.** Regression linear of hydrocracking time vs yield of liquid product

amounts of gasoline and diesel product obtained from hydrocracking of waste lubricant using Ce/MS catalyst were about 8.05 and 12.26 wt.%, respectively. The presence of gasoline and diesel in liquid products from hydrocracking of waste lubricant was proved by the appearance of peaks below a retention time of 20 min on the liquid product chromatogram. The chromatogram of liquid product from hydrocracking of waste lubricant using Ce/MS catalyst is presented in Fig. 7.

Ce/MS Catalyst Lifetime Determination

Table 2 presents product distribution of hydrocracking of waste lubricant using the Ce/MS catalyst from the first to the third run. The amount of liquid product decrease from the first run to the third run. It was caused by deactivation of the catalyst when the catalyst was used repeatedly. The Ce/MS catalyst produced solid product (coke) on the first run of hydrocracking process. This coke was accumulated and blocked the catalyst pore from absorbing the feed on the next run [21].

The Catalyst lifetime was determined using a linear regression between yield of the liquid product obtained from hydrocracking of waste lubricant using the Ce/MS catalyst for three times and hydrocracking time. The regression linear is presented in Fig. 8. The linear regression formula is $y = -2.18x + 27.28$. By using the

regression linear formula, the lifetime of the Ce/MS catalyst for hydrocracking of waste lubricant is 12.54 h. It means that hydrocracking of waste lubricant using the Ce/MS catalyst will not produce liquid product after 12.54 h.

CONCLUSION

The Ce metal was successfully impregnated in the MS synthesized using bovine bone gelatin as a template. The Ce/MS catalyst showed an acidity of 2.79 mmol/g, BJH desorption pore diameter of 3.84 nm, BET surface area of 246.55 m²/g, and total pore volume of 0.44 cm³/g. The yield of liquid product obtained from hydrocracking of waste lubricant using the Ce/MS catalyst for the first, second, and third runs was 21.42, 17.23 and 10.54 wt.%, respectively for 2.5 h per each run. Catalyst lifetime was determined using a linear regression of hydrocracking time versus liquid fraction conversion, resulting the Ce/MS lifetime of 12.54 h.

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