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# Adsorption of ibuprofen molecule onto mesoporous silica SBA-15 loaded by iron particles using arc discharge treatment

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**Abstract.** In this article, adsorption performance of mesoporous silica SBA-15 loaded by iron particle via arc discharge methods has been investigated for the first time using ibuprofen as molecule adsorbate model. The block copolymer poly-ethyleneglycol-block-poly- propylene glycol-block-poly-ethylene glycol P123, tetraethoxysilane and chloride acid has been acted as nano templating, silica source and catalyst during preparation of mesoporous silica SBA-15. The mesoporous silica SBA-15 has been synthesized by hydrothermal technique which is followed by arc discharge iron treatment for adding magnetic character. Transformation of mesoporous silica structure has been investigated by small angle X-ray diffraction (XRD), scanning electron microscopy (SEM) and nitrogen adsorption-desorption techniques, Fourier-transform infrared spectroscopy (FTIR) and elemental analysis. The result showed that magnetic properties mesoporous silica SBA-15 increased after arc discharge treatment without significant structural damages. The ibuprofen adsorption prepared by using 20 mg adsorbent dose and time for contact between adsorbent and adsorbate in 100 min at room temperature. The adsorption capacity of mesoporous silica SBA-15 investigated by UV-Vis show that maximum adsorption capacity of SBA-15 was up to 77.5 mg.g<sup>-1</sup> on optimum condition.

**Keywords:** adsorption, ibuprofen, molecular sieve silica SBA-15, arc discharge, capacity

## 1. Introduction

Many drug such as ibuprofen, paracetamol, and other pharmaceutical product have been released into the aquatic area. Ibuprofen as anti-pyretic (NSAID type) from the specific investigation report, ibuprofen with range concentration 0.018 up to 2.110 µg/L was found in the US, Germany and several aquatic area in the world [1]. This phenomenon has negative impact for digestion system of water organisms due to the faster growth of microorganism. The harmful effect of ibuprofen for aquatic area can destruct the whole ecosystem. For human, ibuprofen could be produce gastrointestinal effect and destructed the brain system [2]. The solution of these problem have been investigated by many researcher during last ten years.

The study for minimize the concentration of ibuprofen in aquatic life was found that biological process by biodegradation have limitation for drinking process. The other method for ibuprofen removal have been proven by researcher such as oxidation, filtering, radiation and adsorption with limitation for each method [3]. Adsorption is still popular and favourable for many drinking industrial need to remove drug compound from aquatic area. The reason of the adsorption popularity is low cost, simple, high



regeneration power and low energy consumption. The common adsorbents have been used in many adsorption cases such as wood, bagasse, activated carbon, fly ash, zeolite, silica, carbon [4]. These adsorbents were dominated by low surface area which has had a bad impact on low adsorption capacity. Today, materials with high surface area have been successfully synthesized by previous researchers such as mesoporous silica, mesoporous carbon, nano silica, nano metal and nano-zeolite. Mesoporous silica SBA-15 is one of the favourable materials for adsorption due to its large surface area, high pore volume, high thermal stability, high pore uniformity and unique structure that improve the material strength [5-7].

However, SBA-15 has a high cost during production. The solution of these problems is to increase the regeneration ability to minimize the total adsorbent during adsorption. The regeneration could be done if the adsorbent has a magnetic character. In this research, we try to synthesize mesoporous silica SBA-15 with a magnetic character. The mesoporous silica SBA-15 with a magnetic character was synthesized by hydrothermal technique followed by arc discharge iron treatment. The characterization of the material was investigated by SEM, EDX, FTIR, and XRD.

## 2. Experiment

### 2.1. Material

Ibuprofen was purchased from Sigma Life Science, deionized water and n-hexane received from Thermo Scientific Barnstead RO NanoPure system. Tetraethyl orthosilicate (TEOS), cetyltrimethylammonium bromide (CTAB), acrylic acid, potassium persulfate, sodium dodecyl sulphate, ammonium nitrate, ethanol and hexane were supplied by Sinopharm Chemical Reagent Co. Ltd.

### 2.2. Mesoporous silica SBA-15 synthesis

The block copolymer poly-ethyleneglycol–block-poly- propylene glycol–block-poly-ethylene glycol P123 was dissolved in HCl solution at room temperature with a stir rate of 200 rpm until a clear solution was found. The clear solution was added to tetraethoxysilane then the mixture was kept in room temperature under stirring for a night. The clear resulting mixture was transferred into a steel reactor followed by a hydrothermal process at 100°C for 24 h. The white slurry was separated, washed with water, dried at 100°C for 2 h then calcined at 550°C in air for 12 h.

### 2.3. Iron Mesoporous silica (Iron/SBA-15) synthesis by arc-discharge method

Iron/SBA-15 is obtained first from the iron oxide electrolysis process (40 to 70 A for 2 h) using 0.375 M NaCl as electrolyte. Chlorine ions have been removed by  $\text{AgNO}_3$  solution on the filtrate followed by a heating process at 250°C for 1 h to obtain a filler. The magnetic material was made from carbon (graphite), silica glue (as a binder) and SBA-15 and ethanol with a mass ratio of 1: 3: 1:0.01 (w/w). A solid graphite tube measuring 50 mm in length with a diameter of 10 mm was used as a cathode. Anode and cathode are cultivated close to a very small distance but not stick. This is done in order to jump electric ions electricity that is marked by sparks. The current and the electrode, both the anode and the cathode, are set up in the beaker glass containing the urea p.a. solution. (Merck) in ethanol. The current used in this method is 10 A. This process takes place from the appearance of sparks to no longer visible in the time between 10-15 min. The solution medium was used in 50% ethanol arc-discharge. The final product was labelled as iron/SBA-15.

### 2.4. Ibuprofen adsorption

Ibuprofen solution was created by mixing the ibuprofen powder into the n-hexane until 100 ppm followed by a stirring process in room temperature for 12 h. A 50 mg adsorbent iron/SBA-15 was mixed with 50 mL ibuprofen solution with a stirrer at 20°C for 1 h followed by sampling 3 mL ibuprofen solution at 0; 5; 10, 15, 20 until 100 min. Ibuprofen concentration measurement after adsorption using UV-visible spectrophotometer was operated at a wavelength of 265  $\text{nm}$ . After sample analysed by the

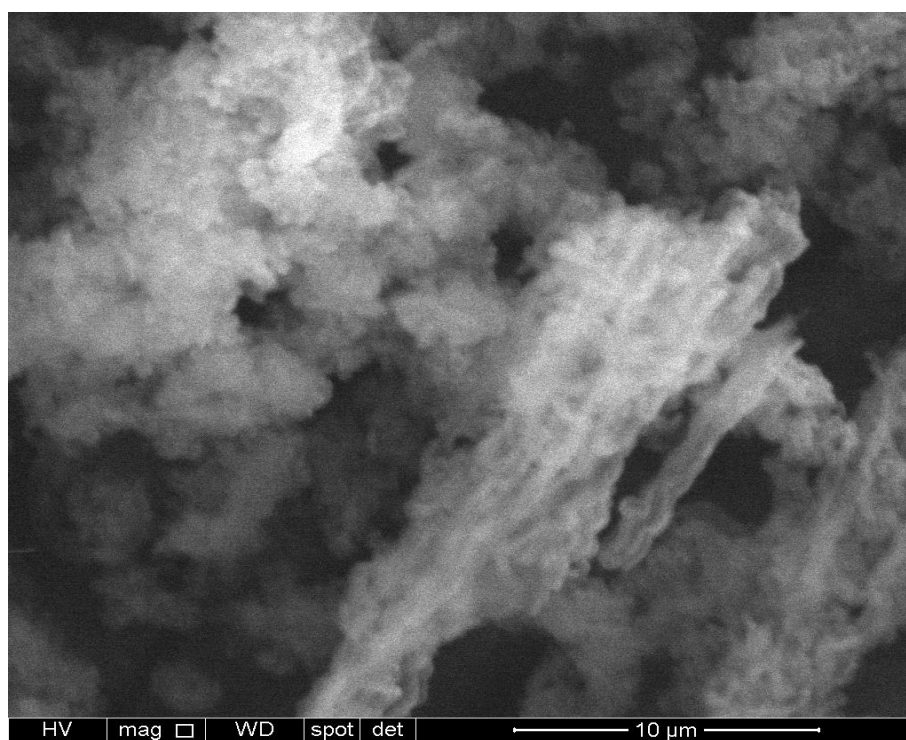
spectrophotometer, the end absorbance value was used to find ibuprofen concentration. In the final step, concentration of ibuprofen solution was used to find adsorption capacity of material.

### 2.5. Characterization

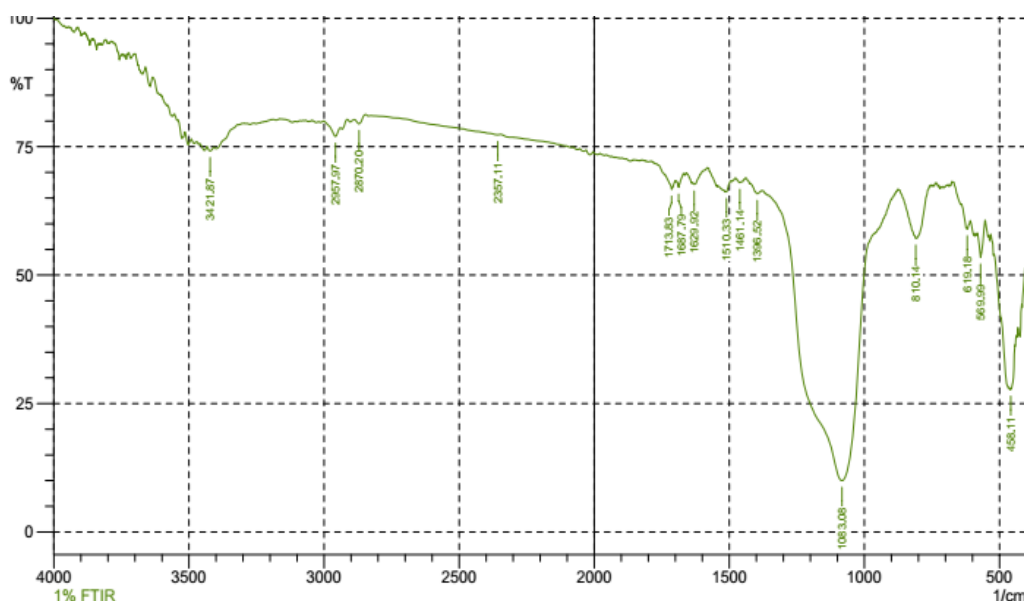
The iron/SBA-15 obtained from the arc discharge were characterized using an X-Ray Diffractometer (XRD-6000, Shimadzu) (Cu; 40.0 kV; 30.0 mA). The morphology of iron/SBA-15 were studied using a scanning electron microscope (JEOL SEM-1400). The functional group of iron/SBA-15 were investigated by FTIR

## 3. Result and Discussion

Fig 1 show morphologies and microstructure of iron/SBA-15. As can be seen in the SEM image, the shape of iron/SBA-15 was observed close to the pipe-like which is have good agreement with SBA-15 in the previous report as parent material. The size of iron/SBA-15 pipe was around 2-10  $\mu\text{m}$ . However, the SEM image clearly displayed the over lapping occurrence among nano pipe of SBA-15 without any significant destruction part by iron infiltration, implying that the mesoporous silica of SBA-15 was completely coated by iron precursor.



**Figure 1.** SEM of SBA-15 by arc-discharge method after ibuprofen adsorption.

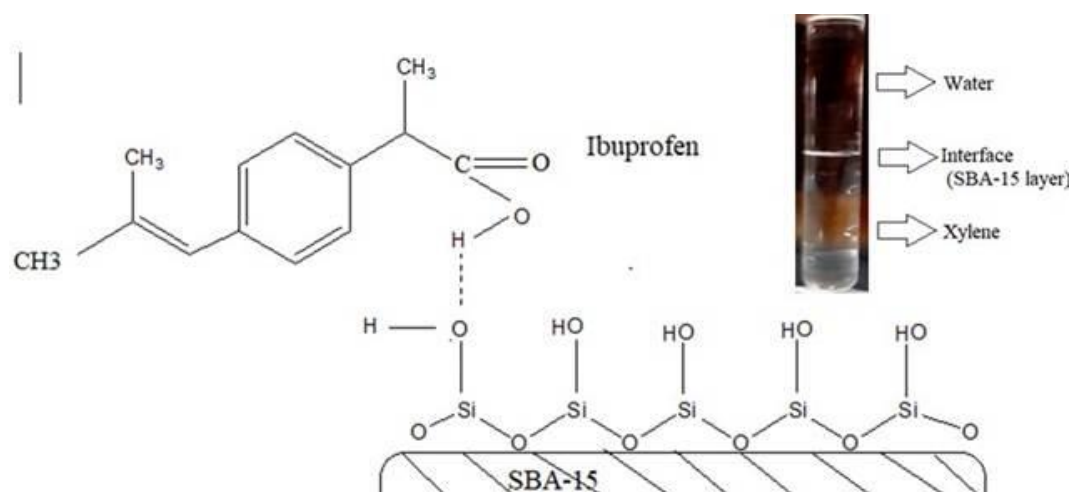


**Figure 2.** FTIR Spectra of SBA-15 y arc-discharge method after ibuprofen adsorption.

Spectra of iron/SBA-15 after ibuprofen loading are shown in Fig 2. the iron oxide was represented by the adsorption band at  $676.08\text{ cm}^{-1}$  and  $1384\text{ cm}^{-1}$ . This means that sample of iron/SBA-15 have iron oxide content after arc-discharge loading effect. The vibration adsorption peak of the silanol Si-OH bond, which was originally located at  $1639, 56\text{ cm}^{-1}$  and siloxane Si-O-Si bond at  $1085\text{ cm}^{-1}$  [8, 9]. Fig. 2, shows the absorption peak of the Si-OH band at  $1635.71\text{ cm}^{-1}$  and Si-O-Si band at  $1084.04\text{ cm}^{-1}$ . The peak of silanol and siloxane suggested that the framework of mesoporous silica SBA-15 was quite stable after all step preparation process. However, the vibration peak of the C=O, C-C and C=C band at  $1461.14, 1510.33, 1687.79$  and  $2957.97\text{ cm}^{-1}$  is appeared which indicated that ibuprofen still covered at iron/SBA-15 after adsorption.

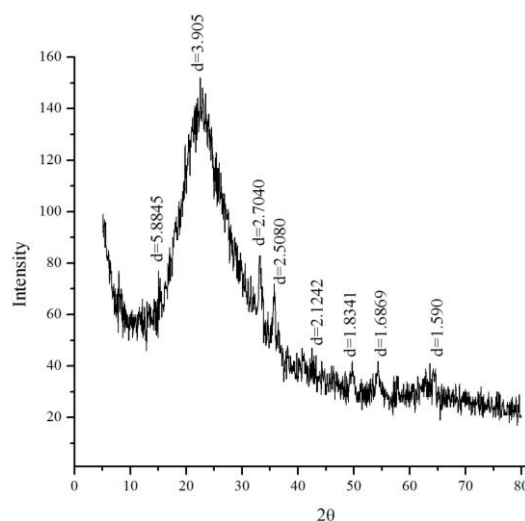
**Table 1.** Functional group adjustment for mesoporous silica SBA-15 after ibuprofen adsorption.

No	Peak	Functional group	Intensity	Corr. Intensity	Area
1	458.11	CH <sub>2</sub> stretching vibration	27.75	0.591	8.067
2	569.99	CH <sub>2</sub> stretching vibration	53.449	5.707	6.735
3	619.18	Fe-O stretching vibration	58.946	2.676	7.103
4	810.14	Si-O-Si stretching vibration	57.162	0.558	13.041
5	1083.08	Si-O-Si stretching vibration	10.00	50.338	204.957
6	1396.52	Fe-O stretching vibration	65.85	0.486	4.335
7	1461.14	C=O stretching vibration	68.004	0.469	3.994
8	1510.33	C=O stretching vibration	66.218	0.145	2.819
9	1629.92	stretching vibration in carboxylic acid group	67.66	0.717	7.306
10	1687.79	C=C stretching vibration	67.076	0.876	2.279
11	1713.83	C=O stretching vibration	66.675	0.639	4.195
12	2357.11	CH <sub>2</sub> stretching vibration	77.316	0.128	4.823
13	2870.2	CH <sub>2</sub> stretching vibration	79.458	1.228	3.889
14	2957.97	CH <sub>3</sub> stretching vibration	77.012	2.012	6.313
15	3421.87	-OH, Si-OH stretching vibration	74.178	0.812	3.331



**Figure 3.** Illustration of Ibuprofen adsorption onto SBA-15 surface (insert: hydrophilicity test of SBA-15 surface).

Fig 3. describe the ibuprofen interaction during adsorption onto Iron/ SBA-15 material. The ibuprofen adsorption prepared by using 20 mg adsorbent dose and time for contact between adsorbent and adsorbate in 100 min at room temperature. The adsorption capacity of mesoporous silica SBA-15 investigated by UV-Vis show that maximum adsorption capacity of SBA-15 was up to  $77.5 \text{ mg.g}^{-1}$  on optimum condition (isotherm curve not shown). This result higher than adsorption capacity of SBA-15 without iron in previous report. The high adsorption capacity of the iron/SBA-15 material could be explained by both the chemical and physical factor. The chemical factor of high adsorption capacity represented by the electrostatic attractions between the carboxylate function of ibuprofen compound and the positive charges present on the silica and iron surface and some  $\pi - \pi$  dispersive interactions between the iron ion and the single carbon layers of ibuprofen chain. The electrostatic attraction also can be seen during hydrophilicity test using water and xylene (Fig 3, inset). More than 30% of iron/SBA-15 part still on the water part and more 50% iron/SBA-15 stable on interface part which is explain that iron/SBA-15 has hydrophilicity character to improve the interaction with ibuprofen during adsorption. The physical factor represented by high surface area of iron/SBA-15 at about  $450 \text{ m}^2/\text{g}$  and the uniform pore shape which is could improve the deposition ibuprofen efficiency.



**Figure 4.** XRD of Iron SBA-15 by arc-discharge method.

Diffraction patterns for iron/SBA-15 are shown in Fig. 4. that SBA-15 showed two typical diffraction peaks on the reflection (0 1 2) and (1 1 0) at  $2\theta$  25.5° and 33.5°. The existence of silica peak after arc discharge process was confirmed that silica part still stables after iron infiltration. The iron oxide of Fe<sub>3</sub>O<sub>4</sub> planes on reflection (220), (311), (400), (422), (511), and (440) in Fig 4 also represented by peaks at 30.5296°; 35.9186°; 43.6172°; 53.9676°; 57.5602° and 74.6690°. The iron oxide also imply that Fe/SBA-15 have magnetic character that can be used by separating agent between filtrate and suspension in aquatic environment. As can be seen, the strength of silica peak and iron peak indicated that iron oxide was completely infiltrated onto mesoporous silica SBA-15 after arc discharging process. Moreover, the existence of iron in this material could be the promising adsorbent candidate due to the magnetic character to improve regeneration efficiency.

#### 4. Conclusion

Mesoporous silica SBA-15 loaded by iron particles have been synthesized using arc discharge treatment. The result show that iron/SBA-15 material have regular mesopore type with pipe-like structure at length range about 2-10.0  $\mu\text{m}$ . X-ray diffraction, scanning electron microscopy FTIR shows that iron SBA-15 sample have been observed as the impact of arc-discharge process. The iron oxide Fe<sub>3</sub>O<sub>4</sub> XRD pattern imply that Fe/SBA-15 have strong magnetic character that in future can be used as separating agent in ibuprofen remediation from aquatic area. The adsorption performance using mesoporous silica SBA-15 loaded by iron particles sample was investigated by ibuprofen adsorption which concluded that electrostatic interaction and surface area are the major factor to enhance adsorption capacity.

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