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INFLUENCE OF TYPE OF SUPPORT ON ADSORPTION OF
MERCURY COMPOUNDS FROM A LIQUID HYDROCARBON
USING COPPER OXIDE AS AN ACTIVE METAL



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สิบทิศ ประสมศักดิ์: ผลของชนิดของตัวรองรับต่อการดูดซับสารประกอบปรอทจากไฮโดรคาร์บอนเหลว โดยการใช้โลหะว่องไวชนิดคอปเปอร์ออกไซด์ (INFLUENCE OF TYPE OF SUPPORT ON ADSORPTION OF MERCURY COMPOUNDS FROM A LIQUID HYDROCARBON USING COPPER OXIDE AS AN ACTIVE METAL)

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การวิจัยครั้งนี้ เป็นการศึกษาผลของตัวรองรับชนิดอะลูมินา ซิลิกา ถ่านกัมมันต์ ซิลิกา-อะลูมินาและไททาเนียมออกไซด์ ต่อการดูดซับสารประกอบปรอทจากไฮโดรคาร์บอนเหลวโดยใช้โลหะว่องไวชนิดคอปเปอร์ออกไซด์ ปริมาณคอปเปอร์บนตัวดูดซับคอปเปอร์ออกไซด์ร้อยละ 2.5 โดยน้ำหนัก ทำการทดลองที่ความดันบรรยากาศ ที่อุณหภูมิ 30 50 และ 75 องศาเซลเซียส ใช้เมอคิวริกคลอไรด์เป็นตัวแทนสารประกอบปรอทในรูปของโลหะอินทรีย์และใช้ไดฟีนิลเมอคิวรีเป็นตัวแทนสารประกอบปรอทในรูปของโลหะอินทรีย์ สารตั้งต้นประกอบด้วยสารประกอบของปรอทละลายในโทลูอีนที่มีความเข้มข้นของปรอท 1 ส่วนในล้านส่วน

จากผลการทดลองพบว่าตัวดูดซับคอปเปอร์ออกไซด์มีประสิทธิภาพในการดูดซับปรอทออกจากไฮโดรคาร์บอนเหลวได้ดี ความสามารถในการดูดซับสารประกอบปรอทของตัวดูดซับขึ้นกับอุณหภูมิและชนิดของสารประกอบปรอท โดยตัวดูดซับทั้งหมดสามารถกำจัดเมอคิวริกคลอไรด์ได้ดีกว่าไดฟีนิลเมอคิวรี ความสามารถในการดูดซับสารประกอบปรอทยังขึ้นกับชนิดของตัวรองรับ ความสามารถในการดูดซับสารประกอบปรอทของตัวดูดซับคอปเปอร์ออกไซด์บนตัวรองรับแต่ละชนิดจะเพิ่มขึ้นตามลำดับดังนี้ ถ่านกัมมันต์ > อะลูมินา > ซิลิกา-อะลูมินา > ไททาเนียมออกไซด์ > ซิลิกา เมื่อสารประกอบปรอทถูกดูดซับบนตัวดูดซับแล้วเกิดสารประกอบคอปเปอร์เมอร์คิวรี (CuHg และ Cu₁₅Hg₁₁) บนตัวดูดซับคอปเปอร์ออกไซด์

ภาควิชา.....วิศวกรรมเคมี.....ลายมือชื่อผู้ผลิต.....
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Influence of type of support on adsorption of mercury compounds from liquid hydrocarbon was investigated in this study. The adsorbents were copper oxide on different types of supports. The supports used in this study were alumina, silica, activated carbon, silica-alumina and titanium oxide. Copper loading was 2.5% by weight. The experiments were conducted at atmospheric pressure and at temperatures of 30°C, 50°C and 75°C. Mercuric chloride and diphenylmercury were used to represent inorganic and organic forms of mercury compounds. These compounds were dissolved in toluene to obtain solutions containing 1 ppm of mercury.

The results showed that copper oxide adsorbents could be used effectively in the removal of mercury compounds from liquid hydrocarbon. Removal of mercury depended on operating temperature. In addition, it also depended on the types of mercury compounds. The results showed that mercuric chloride could be removed more effectively than diphenylmercury by all adsorbent. Removal of mercury also depended on types of supports. Adsorptive ability of adsorbent on mercury removal was in the following order: copper oxide/alumina > copper oxide/activated carbon > copper oxide/silica-alumina > copper oxide/titanium oxide > copper oxide/silica. Formation of copper mercury (CuHg and $\text{Cu}_{15}\text{Hg}_{11}$) was detected on spent copper oxide adsorbents.

Department.....Chemical..Engineering.....Student's signature.....
Field of study...Chemical..Engineering.....Advisor's signature.....
Academic year.... 2001.....Co-advisor' signature.....

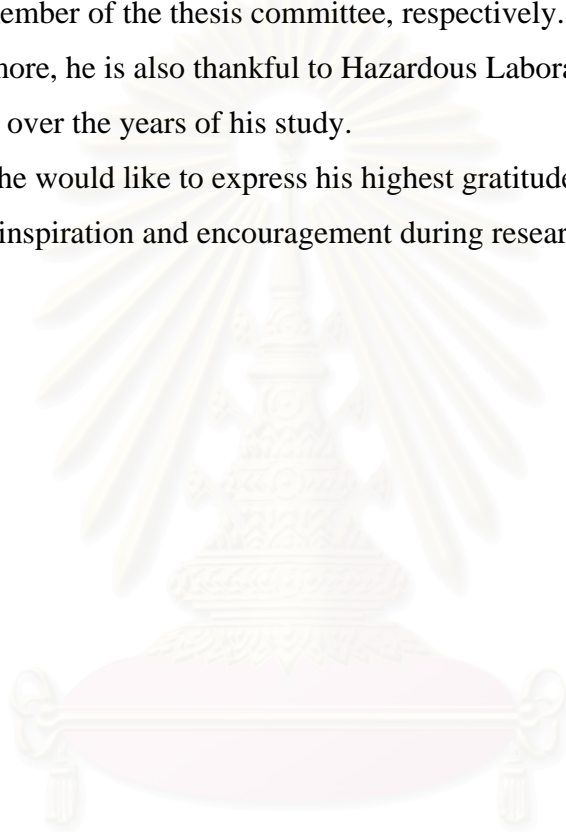
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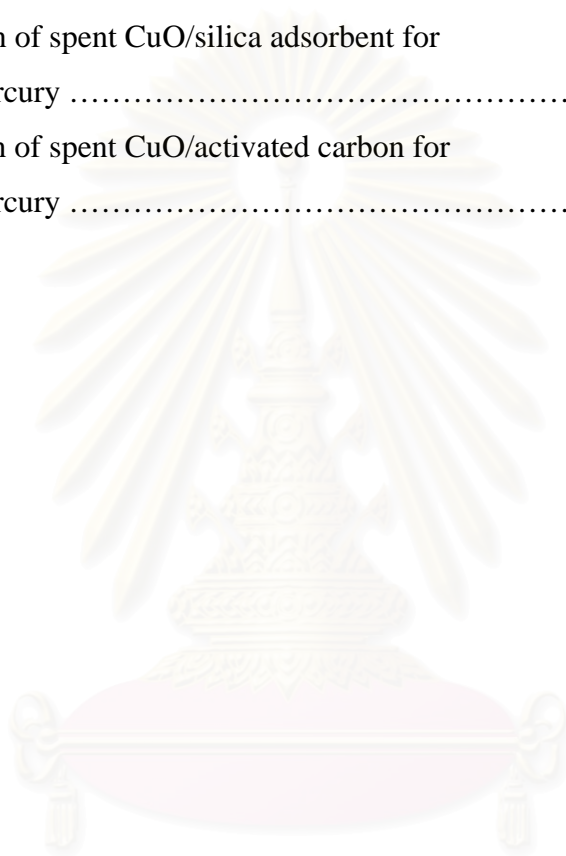
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CHAPTER I

INTRODUCTION

Petroleum, which is used as feedstock for refinery and petrochemical process contains hydrocarbon as main composition and some impurities. These impurities are usually found in the form of sulfur, nitrogen, oxygen, and metal compounds. Mercury is one of metal impurities that can be found.

The concentration of mercury in liquid hydrocarbon is highly dependent on geologic location and varies between approximately 0.01 ppb and 10 ppm (wt.)(Wilhelm and Bloom, 2000). Various mercury species are detected in liquid hydrocarbon such as elemental mercury, inorganic and organic mercury compounds.

Although mercury found in trace quantity, it causes problems in processing due to the corrosive effect of mercury on vital equipments such as cryogenic heat exchanger. In addition, catalyst used in catalytic processes such as catalytic hydrogenation is susceptible to mercury poisoning.

Several methods have been proposed for mercury removal from liquid hydrocarbon. It can be classified into two groups, chemical treatment and adsorption. Chemical treatment is the method that using chemicals to convert mercury to the form which is easy to remove. The other method, adsorption, is the most widely used for mercury removal. This method is the contacting liquid hydrocarbon with an adsorbent under suitable conditions. The adsorption method provides a high efficiency of mercury removal.

Many types of adsorbent are proposed for removal of mercury from liquid hydrocarbon. The adsorbent comprises of two parts, active metal and support.

There are many types of active metal used in the mercury removal processes. Torihata and Nisimura (1989) studied a process to removing mercury from H-NGL (heavy natural gas liquid) by used ferric oxide (Fe_2O_3), copper oxide (CuO), and nickel oxide (NiO) on alumina as the adsorbent. They found that CuO is more effective in mercury removal than Fe_2O_3 and NiO . Yan (1990) studied a method for removing mercury from heavy hydrocarbon condensate by high temperature reactive adsorption. The adsorbents that used in the process were $\text{CuO}/\text{Al}_2\text{O}_3$, $\text{CuS}/\text{Al}_2\text{O}_3$,

Fe/Al₂O₃, and Ag/Al₂O₃. He found that CuO is more effective in mercury removal than CuS, Fe, and Ag. Tantichaipakorn (1998) was found that copper oxide adsorbent is more effective than nickel oxide adsorbent.

There are many solid materials are used as support such as alumina (Torihata and Nisimura, 1989), silica (Kawazoe, 1990), activated carbon (Yan, 1996), silica-alumina (Yan, 1987) and titanium oxide (Sugier et. al., 1978). Kawazoe (1990) found that the use of different supports would effect to efficiency of mercury removal process. Yan (1996) found that activated carbon-based adsorbent is more effective than the alumina-based adsorbent.

The objectives of this research are to study the influence of support on the adsorption of mercury compounds and to determine the effect of operating temperature on mercury removal. In the present study, alumina, silica, activated carbon, silica-alumina and titanium oxide are used as supports of 2.5% copper oxide adsorbents. Toluene containing mercury compounds is used as the feed model. Mercuric chloride and diphenylmercury are selected as mercury compounds in inorganic and organic forms respectively. Liquid product is digested with permanganate solution, persulfate solution, nitric acid and sulfuric acid which conform to ASTM D-3223 to obtain the inorganic form in aqueous phase before analyzed by atomic absorption spectrometry. Total surface area, pore volume, average pore diameter and micropore area of each supports and copper oxide adsorbents are analyzed by BET method.

CHAPTER II

LITERATURE REVIEWS

2.1 Mercury compounds in petroleum

Mercury is usually found in a wide range of petroleum such as natural gas, condensate and crude oils. Quantities of mercury depend on the sources of petroleum feedstocks. Natural gas and associated condensate are found to have mercury contents of 15 to 450 ppb (Yan, 1987) and 10 to 3000 ppb (Sarrazin, 1993), respectively. Low level of mercury can be found in crude oils at range of 0.5 to 10 ppb (Yan, 1990).

Mercury in natural gas is generally an elemental form. Natural gas condensate is different. Various mercury species are detected in condensate such as elemental mercury, inorganic and organic mercury compounds. Inorganic, HgCl_2 , and organic, $\text{Hg}(\text{CH}_3)$, $\text{Hg}(\text{C}_2\text{H}_5)_2$, $\text{Hg}(\text{CH}_3)\text{Cl}$, forms are found more often than elemental mercury. The solubility of some mercury compounds has been reported by Wilhelm (1999b) (Table 2.1)

Table 2.1 Approximate Solubility of Mercury Compounds at 25°C (Wilhelm, 1999b)

	Water(ppb)	Oil(ppb)	Glycol(ppb)
Hg^0	50	2,000	7,000
HgCl_2	70,000,000	>10,000	>50,000
HgS	10	Very low , <10	Very low , <10
HgO	50,000	Low	
CH_3HgCl	Very high	1,000,000	High

2.2 Disadvantages of mercury compounds in petroleum

2.2.1 Plant corrosion by mercury

High levels of mercury in hydrocarbon liquid, crude oil and condensate can cause problems in processing due to the corrosive effect of mercury on vital equipment such as cryogenic heat exchanger. Such that exchangers are often

made from aluminum which forms an amalgam with mercury, for example, failures occurred at the LNG plant at Skikda, Algeria, from tube corrosion in the spiral wound exchangers (Leeper, 1980). Corroded tubes contained white deposits: aluminum oxide, aluminum hydroxide and aluminum carbonate, with traces of elemental mercury.

2.2.2 Catalyst deactivation

Most catalysts used in catalytic processes may lose their activity or their selectivity when it is used for a long time. It is known in the term of “Deactivation”. The causes of catalyst deactivation may be divided into four groups: poisoning, fouling, sintering and loss of active species. Catalyst poisoning may result from an impurity containing in the feed. The impurity adsorbs on active sites of catalyst and reduces catalyst activities. Fouling is generally used to describe a physical block such as the deposit of dust or coke. Sintering is an irreversible physical process leading to a reduction of catalyst activity. Finally, is less active or selective. Deactivation of catalyst that is caused by mercury is grouped into the chemical poisoning.

For reforming hydrocarbon oils such as naphtha by, for example, hydrogenation, such catalysts as palladium catalyst support on alumina are used. On the other hand, if mercury and its compounds are present in hydrocarbon oils as impurities, such reaction as hydrogenation cannot be carried out sufficiently due to catalyst poisoning caused by such impurities (Torihata, 1989).

In addition, mercury compounds are extremely toxic with man and animals, especially organic compounds. They damage nervous system on inhalation or ingestion.

2.3 Methods for eliminate mercury compounds from liquid hydrocarbon

Several methods have been conventionally practiced for removal of mercury and its compounds from liquid hydrocarbon. The methods can be classified into two groups: chemical treatment and adsorption.

2.3.1 Chemical treatment

Chemical treatment is a method that converts mercury in liquid hydrocarbon by reacting with some chemicals. The chemical used is usually a sulfur compounds, such as alkali polysulfide. The reaction between sulfur and mercury is shown below.



The occurred mercuric sulfide, HgS, is a solid material that cannot dissolve in hydrocarbon and is removed easily from the feed stream. By this method, natural gas condensate is contacted with aqueous solution of sulfide in several different ways, both in batch-wise or continuous processes.

Yan (1988) proposed a method for removing mercury from natural gas condensate. The method comprises contacting the contaminated condensate with dilute aqueous solution of alkali metal sulfide salt, Na_2S_x . The mercury content in the condensate was 220 ppb. The study was carried out by mixing the condensate with Na_2S_x and aqueous NaOH solution of varied concentration at temperature of 75°C . After homogenization, the mixture was allowed to settle for 1-5 minutes. The result shows that the important variables in removing from the condensate are intensity of mixing, concentration of Na_2S_x , volume ratio of caustic solution of Na_2S_x , and efficiency of phase separation. For example, Na_2S_x solution containing 2000, 1000, 500 and 100 ppm of sulfur, each containing 0.8 wt% of NaOH was mixed with the condensate. The mercury concentration decreased correspondingly from 220 ppb to 66, 133, 77 and 110 ppm, respectively. The amount of mercury removed increases with an increase in sodium sulfide concentrations but it does not proportionally improve mercury removal. To study the effect of mixing intensity, the blender was set and connected to a power-stets, which was varied from 20 % to 100 %. At 20 and 30 % setting, the mercury content in product was decreased to 180 ppb. As the power setting was increased to 50 and 100 %, the removal efficiency increased and the mercury concentration in the products decreased to 134 and 71 ppb, corresponding to 33 and 65 % mercury removal. These results show the effect of mixing intensity for Hg removal. A residual mercury containing in liquid hydrocarbon was removed by mixing with aqueous polysulfide solution (Audeh, 1989). In this process, pentane containing 13 ppb of mercury was mixed with approximately 0.5 cc of sodium

polysulfide which contained 22.2 wt% of sulfur. The process was carried out at ambient or room temperature of 70°F. After treating, the mercury content in product was less than 0.01 ppb. However, the blank was tested by the same procedure except mixing the hydrocarbon with sulfide solution. It was found that mercury content did not change but remained at 13 ppb.

2.3.2 Adsorption

The removal of mercury by adsorption is an extractive method. It provides a high efficiency of Hg removal and does not contaminate with other chemical. The adsorption method comprises contacting the hydrocarbon with an adsorbent at various conditions, depending on type of adsorbent used. Mercury is adsorbed and remains in the adsorbent. Thus, the treated hydrocarbon is readily free from mercury contamination.

Mercury removal adsorbent beds are used to scavenge mercury from liquid hydrocarbon streams. The adsorbents consist of granular or palletized material consisting of a substrate support (activated carbon, metal oxide or alumina) and a active metal (CuO, CuS, etc.) that is bonded to the support.

There are many studies about active metal.

Sugier et al.(1978) studied a process for removing mercury from a gas and a liquid hydrocarbon. The adsorbent used was reduced copper and copper sulfide on alumina and cement (70% Al₂O₃, 26% CaO, 0.5% Na₂O, 0.2% SiO₂ and 3.3% Miscellancous). The condition was 3500 kPa and ambient temperature. The result found that the treat hydrocarbon had mercury below 10 % of fresh feed.

Leeper (1980) proposed corrosion of LNG plant caused by mercury and also method for removal of mercury. For example, natural gas contaminated with mercury was contact with a fixed bed of metal sulfide on silica-alumina support.

Yan (1987) suggest a process for removing mercury from liquid hydrocarbons by adsorption. The adsorbent used was reduced bismuth on alumina (Bi/Al₂O₃). Hexane containing 160 ppb Hg was passed through the adsorbent. At 100°F, the Hg content in hexane was reduced to 1 ppb.

Torihata and Nisimura (1989) studied a process to removing mercury from H-NGL (Heavy natural gas liqiud) by used Fe₂O₃, CuO, and NiO on alumina as the

adsorbent. At 200°C the result found that Fe₂O₃/Al₂O₃ removed mercury from 150 ppb to 18 ppb, CuO/Al₂O₃ removed to 8 ppb and NiO/Al₂O₃ removed to 13 ppb.

Yan (1990) studied a method for removing mercury from heavy hydrocarbon condensate by high temperature reactive adsorption. The adsorbent used was CuO/Al₂O₃, CuS/Al₂O₃, Fe/Al₂O₃ and Ag/Al₂O₃. Condensate contains 220 ppb Hg was passed through the adsorbent at 450°F. The remaining concentration was 14, 51, 124 and 19 ppb, respectively.

Ou (1995) studied a method for removal of mercury by using an adsorbent. This method was directed to an effective way of removing elemental and inorganic mercury from liquid hydrocarbon. The adsorbent used was reduced copper on zinc oxide and alumina that performed virtually removed all mercury in condensate feed. Another adsorbent used was reduced nickel on clay, which reduced 90% mercury of Algerian condensate containing 32 ppb of mercury.

Yamada et al.(1995) studied a mercury removal process for natural gas condensate. They found that mercury compounds, especially organic mercury compounds, could not be adsorbed on any type of adsorbent and not be extracted with any type of agent. However, they found that the mercury compounds could be easily decomposed and converted into elemental mercury by catalyst.

Tan et al. (1996) proposed a method for removal of organic mercury compound from hydrocarbon fraction by heat treatment. High-temperature heat treatment was used to convert an organic mercury compound into an inorganic mercury compound or elemental mercury at a temperature about 200°C to 900°C. Then hydrocarbon fraction was contacted with an adsorbent in the form of active carbon having at least one of calcium and a calcium compound supported there on. The adsorbent was efficient to remove mercury and cost low capital on an industrial large scale, achieving an extremely low mercury concentration.

Sookkho (1995) studied the removal of mercury compounds by adsorption on Cu-Zn adsorbent. His experiments were conducted at 30°C to 75°C and pressure of 200 psig. Mercuric chloride was used as mercury compounds in inorganic form. Phenylmercury acetate and diphenylmercury used as mercury compounds in organic forms. Experimental resulted showed that removal of mercury was significantly dependent on temperature but independent of pressure. In addition, it was also depended on the nature of mercury compounds types.

Tantichaipakorn (1998) studied the removal of mercury compounds by adsorption on CuO and NiO on alumina adsorbent. His experiments were conducted at 30°C to 75°C and pressure of 1 ATM. Mercuric chloride was used as mercury compounds in inorganic form. Diphenylmercury was used as mercury compounds in organic forms. Experimental results showed that removal of mercury was significantly dependent on temperature. In addition, it was also dependent on the nature of mercury compounds types.

Many type of supports are proposed for used in mercury removal process such as alumina (Sugier 1978, Torihata and Nisimura 1989), silica-alumina (Leeper 1980), and activated carbon (Yan 1991). In addition, there are the related research studying about the usage of various supports.

Kawazoe (1990) used many supports such as activated carbon, alumina, silica gel and zeolite. CuCl₂ and SnCl₂ were used as active metal. The liquid hydrocarbon contains 6 ppb Hg was passed through the adsorbent at 10°C. The remaining concentration was below 1 ppb.

Yan (1996) studied a process for removing mercury from oil by adsorption. The adsorbent used was CuO/Al₂O₃, CuS/Al₂O₃, Ag/Al₂O₃ and CuS/activated carbon. The condition was 220 psi and 400°F to 500°F. The result found that the efficiency of the process depended on temperature and type of support. The results showed that the adsorbent, which used activated carbon as the support, could adsorb mercury more than the adsorbent, which used alumina as the support.

2.4 Literature Summary

1. Mercury in petroleum is in elemental, inorganic and organic forms.
2. Mercury present in petroleum leads to corrosion and catalyst poisoning.
3. Methods for mercury removal are classified into two methods: chemical treatment and adsorption. Adsorption is the most widely used method because it provides high efficiency on mercury removal and more convenient.
4. The study of effect of support type on the adsorption of mercury is not clearly known
5. There are many active metals used for removing mercury such as copper oxide, copper sulfide, nickel oxide, nickel sulfide and silver. Copper oxide is preferred.

6. There are many supports used for removing mercury such as alumina, activated carbon, SiO_2 , TiO_2 and silica-alumina.
7. The efficiency of the mercury removal process was independent of pressure but strongly dependent on temperature.



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CHAPTER III

EXPERIMENTS, ANALYSIS TECHNIQUES AND EXPERIMENTAL ERROR

This chapter is divided into four sections: adsorbent preparation, the experimental apparatus and experimental procedure, analytical techniques, and experimental errors.

3.1 Adsorbent preparation

Adsorbents used in the experiment are divided into two groups. The first is solid substances that used as support. In this study, they are alumina, activated carbon, silica, silica-alumina, and titanium oxide. The second is 2.5% copper oxide on these supports. The preparation of adsorbents has two steps. The first is impregnation and the last is calcination.

3.1.1 Impregnation

The copper oxide adsorbents were prepared by wet impregnation of supports with solution of copper nitrate. The apparatus for impregnation is shown in Figure 3.1.

Impregnation procedure

1. Approximately 10 grams of support is added to 250-ml conical flask.
2. The flask is then evacuated, kept pressure at approximately 20 mmHg and heated at 120°C for 3 hours.
3. The flask is cooled to room temperature.
4. Drop distilled water in amount of pore volume of support into the flask under vacuum pressure to fill up pore of support. Vigorously shake the flask for well mix between support and distilled water.
5. Dissolve 0.95 g of copper nitrate in 10 ml of distilled water and then added the solution into the flask for covering the support's surface.
6. The impregnated support was maintained at vacuum pressure for 30 minutes and then adjusted to atmospheric pressure.

- The suspension is then stirred at 300 rpm and 80°C until almost dry and kept at 120°C about 12 hours.

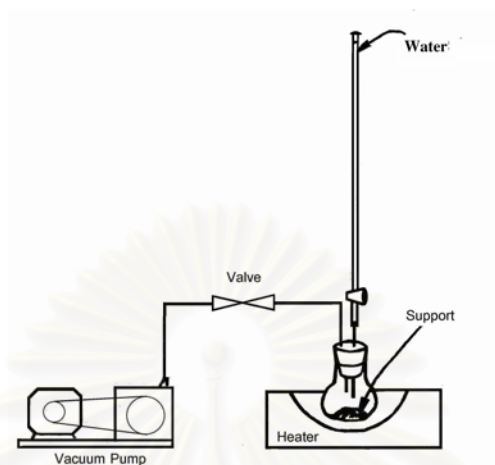


Figure 3.1 Schematic diagram of apparatus for impregnation.

3.1.2 Calcination

Calcination is the method to change forms of copper metal on adsorbent. The calcination is divided into two steps, reduction and oxidation. The first, reduction, copper nitrate is reduced to copper by hydrogen gas. The second step, oxidation, copper is oxidized to copper oxide by air zero. The apparatus for calcination is shown in Figure 3.2.

Calcination procedure

- Approximately 10 g of impregnated support was put in ceramic crucible and placed in pyrex tube following placed in muffle furnace.
- Hydrogen gas was allowed to flow through pyrex tube at a flow rate of 12 l/hr.
- The temperature of the furnace was increased at the rate of 1 °C/min from room temperature to 300°C and maintained at 300°C for 4 hour.
- Air zero was allowed to flow through pyrex tube at a flow rate of 12 l/hr at 300°C and maintained at 300°C for 4 hour.

5. Cool to room temperature in nitrogen flow before the adsorbent was taking out and kept it in the desiccator before use in an experiment.

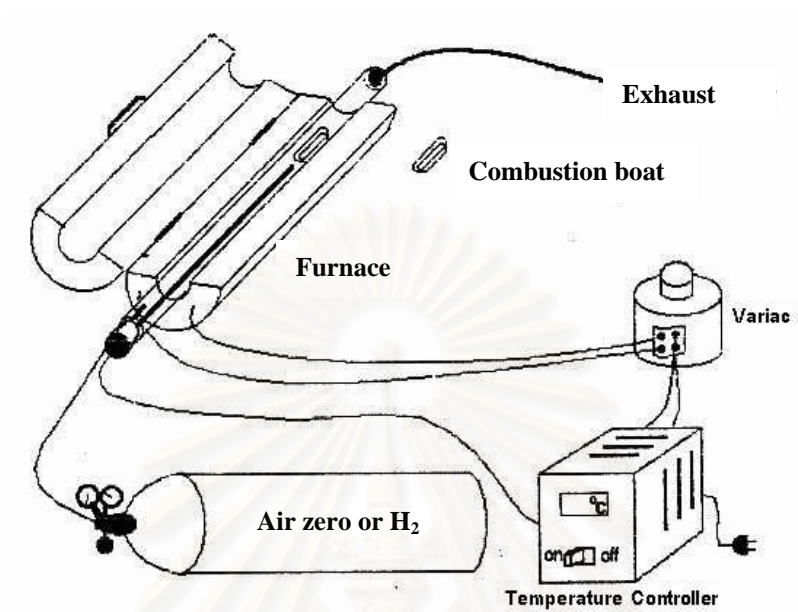


Figure 3.2 Schematic diagram of apparatus for calcination.

3.2 Experimental apparatus and experimental procedure

3.2.1 Experimental apparatus

Figure 3.3 shows schematic diagram of apparatus for experiment. The apparatus are composed of heater, oil bath, temperature controller, motor and propeller. The experiments are carried out at atmospheric pressure. The temperature of oil bath is controlled with temperature controller for 30°C, 50°C and 75°C. The sample is agitated by motor and propeller.

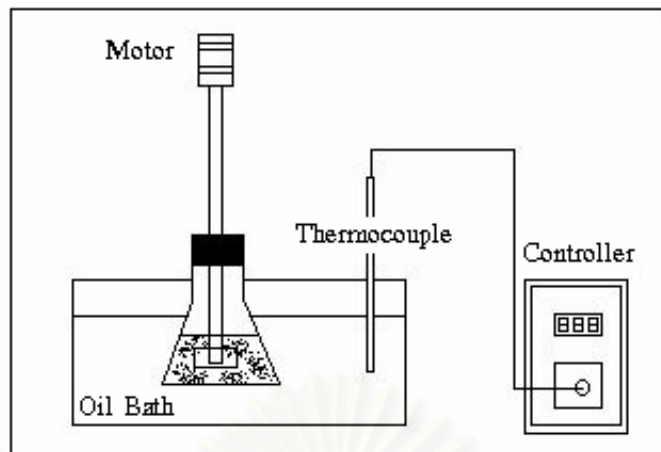


Figure 3.3 Schematic diagram of apparatus for experiments.

3.2.2 Experimental procedure

1. Approximately 1 gram of adsorbent was put into 250-ml flask.
2. The flask was connected to 10-ml burettes and vacuum pump.
3. The adsorbent was filled up by toluene in the amount that equal to pore volume of adsorbent.
4. Approximate 100 grams of liquid feed were put into the flask. The flask was placed into the oil bath that controlled temperature.
5. The experiment was taken place about 60 minutes with stirring at constant temperature.
6. After each experiment, the products were filtrated by Whatman No.1 filter papers to separate the spent adsorbents from liquid product. The adsorbents and liquid solution were separated and both samples were analyzed.

3.3 Analysis Techniques

3.3.1 Adsorbent analysis

Adsorbents are analyzed to specify some characteristics such as percent of metal loaded, form of metal loaded, surface area, pore volume and average pore size

diameter. Techniques and procedure for adsorbent analyses is shown below. The analytical results are shown in chapter IV.

Copper content

Copper content of the adsorbents was determined by standard method of ASTM (base on designation: D1977-91). This method was used for decomposing and dissolving samples for atomic absorption spectroscopy.

1. Approximately 500 mg of the adsorbent sample was weighed and transferred into a crucible.
2. The sample was added with 10 ml of distilled water, 10 ml of concentrated sulfuric acid, 10 ml of concentrated nitric acid, and 5 ml of concentrated hydrofluoric acid.
3. The mixture in crucible was placed on a hot plate and stirred slowly. The adsorbent was first dissolved in acid solution and then precipitated again after the acid was evaporating. A sample was continually evaporated to near dryness.
4. Then the crucible was removed from the hot plate and allowed to cool at room temperature.
5. The sample was introduced again with 20 ml of 19% hydrochloric acid and 30 ml of 3% hydrogen peroxide, covering with watch glass and returning crucible to the hot plate.
6. The sample was continually heated to boil until the precipitate was dissolved totally.
7. After complete dissolution, the crucible was removed from the hot plate and cooled to room temperature.
8. The sample was then diluted with distilled to 100 ml and stored in polyethylene bottle.

Form of metal oxide on support surface

X-ray diffraction technique can determine chemical composition on the surface of material both quantity and quality. The composition occurred on adsorbents are necessary to form sufficient crystalline content for diffracting of x-ray beam.

Adsorbents are prepared to form high content of metal so that x-ray diffraction can detect metal oxide form. The results and patterns of metal oxide adsorbents are shown in chapter IV.

Surface area, Pore volume and Pore size distribution

Surface area and pore characteristics of the samples were measured by the BET method, with nitrogen as the adsorbent using a micromeritics model ASAP 2000. The ASAP 2000 system consisted of two sample preparation ports and one sample analysis ports.

Approximate 100 mg of the adsorbent was weighted and transferred into the sample preparation tube. Most solid adsorbents adsorbed moisture and other contaminants when used. The adsorbent must have been cleaned in sample preparation tube by thermal treating before analysis was performed. The sample preparation tube was attached to the vacuum system and placed around by the heater. Sample preparation would then require more time to achieve the desiring condition before proceeding with an analysis. Once sample preparation was completed, the sample tube might be allowed to cool to ambient temperature. The sample tube would then remove from the sample preparation port and placed onto the analysis port.

Spent adsorbent characterization

Spent adsorbent samples are analyzed to identify mercury content by using method of determining metal content in adsorbents as indicated above. Although the surface area, pore volume and pore size distribution are analyzed by the given method above.

Mercury compound via metal oxide form is analyzed by x-ray diffraction technique. The detection limit is quite high for x-ray diffraction technique with the result that adsorbent has been repeatedly adsorbed by high concentration mercury compound to form high content of mercury compound via metal oxide form. The x-ray diffraction technique's detail is indicated above.

3.3.2 Oil analysis

Mercury digestion from oil

Figure 3.4 shows schematic diagram of the apparatus for digestion. It comprises of 250 ml boiling flask, condenser, oil bath, heater and controller.

Digestion procedure

1. Approximately 30 g of sample was transferred to 250 ml boiling flask.
2. After that 5 ml of concentrated sulfuric acid and nitric acid were added and mixed after each addition.
3. Then, 15 ml of potassium permanganate solution was added to each flask. The mixture was stirred vigorously for at least 15 min.
4. Then, 8 ml of potassium persulfate was added to the flask.
5. The flask at the top was equipped with a reflux condenser and subsequently heated in oil bath at 95 °C for approximately 2 hours.
6. After that the flask cooled to ambient temperature, and added 6 ml of sodium chloride-hydroxylamine hydrochloride solution.
7. Then, shaken for a few seconds. The solution was transferred into 250 ml separating funnel and shaken vigorously.
8. After that, acid-phase was separated from toluene-phase. The remaining toluene-phase was extracted by water for transferring mercury to water phase. The acid-phase and the extracted water were mixed and made the total volume to 100 ml.

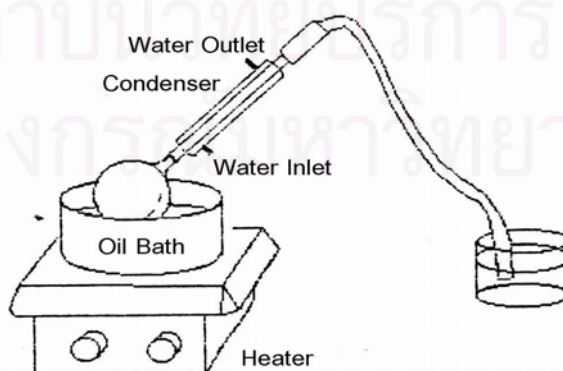
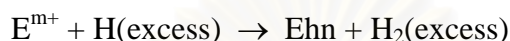
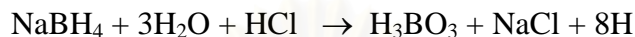


Figure 3.4 Schematic diagram of apparatus for digestion.

Mercury content

Flow Injection Mercury Hydride System Analysis is a high sensitivity and suitable technique for measurement of mercury. The hydride technique involves the reaction of acidified aqueous samples with a reducing agent such as sodium borohydride. The sodium borohydride/acid reduction generates hydrides as shown in the following equations



where E = the analyze of interest and m may or may not equal n

This reaction generates a volatile hydrides which was transported to a quartz cell by argon carrier gas. In the quartz cell, the hydrides were converted to gaseous metal atoms. Although this technique is suitable for measuring of mercury in water-phase, it is applicable to the analysis of materials other than water-phase if and only if, an initial procedure for digesting and oxidizing the sample is carried out. Digestion and oxidization was performed to ensure that the mercury in the sample was converted to the mercuric ion, and dissolved in aqueous media. The digestion method used in this study is applied from ASTM D-3223 which is a standard method for determining of total mercury in water.

3.4 Experimental error

3.4.1 Blank test

The experiments in this section are conducted to study the stability of each metal compound at adsorption temperature and to verify adsorption of metal compounds on glassware wall. No adsorbent is used in this test. The concentration of mercury product is plotted with the operating temperature and shown in Figure 3.5. Percent loss from feed at various temperatures is summarized in Table 3.1. This indicates that all model compounds does not adsorb on the glassware and does not disappear by other parameters. The difference of concentration between feed and product is considered as the error in analysis.

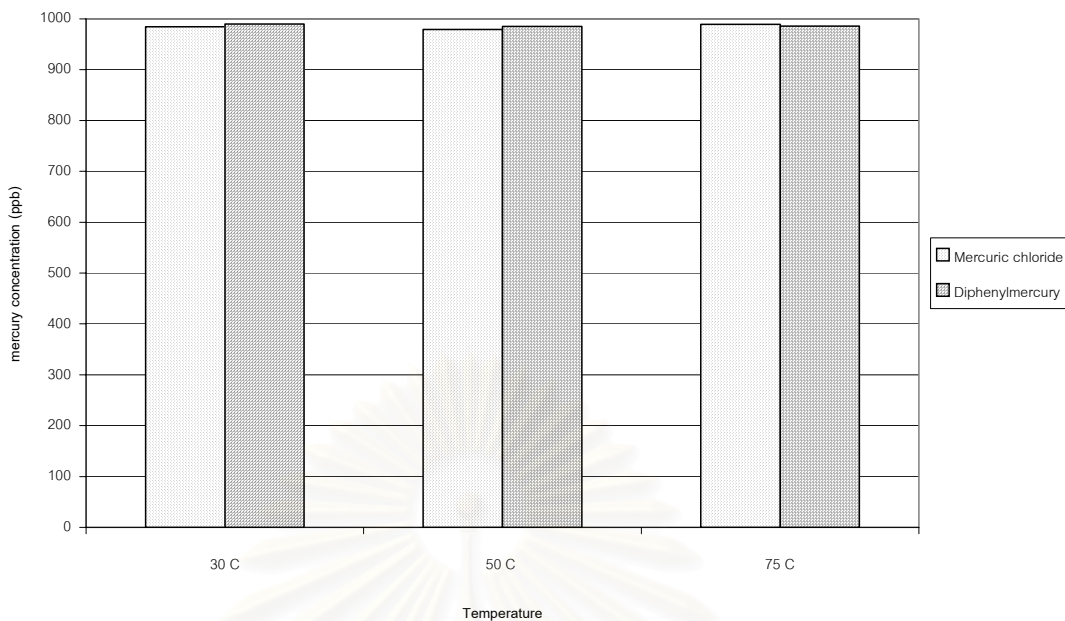


Figure 3.5 Remaining of Mercury in the Study on Blank Test at Various Temperatures

Table 3.1 Percent Loss from Feed at Various Temperatures.

Component	Percent loss from feed (%)		
	30°C	50°C	70°C
Mercuric chloride	1.57	2.08	1.04
Diphenylmercury	1.00	1.50	1.45

3.4.2 Instrumental error

Error from repeated experiments

Some experiments are repeated to study error that might occur during each experiment. The experiments are operated at various temperatures of 30, 50 and 70°C, and adsorbent used is CuO/alumina adsorbent. In addition, both mercuric chloride and diphenylmercury, at initial concentration 1000 ppb, are represented in the experiments. The weight of adsorbent used is 1.0 gram for each experiment. The experiment is repeated for 5 times at the same temperature and mercury compound. Experimental results are illustrated in Figure 3.6 to 3.7. Average concentration and deviation data are shown in Table 3.2.

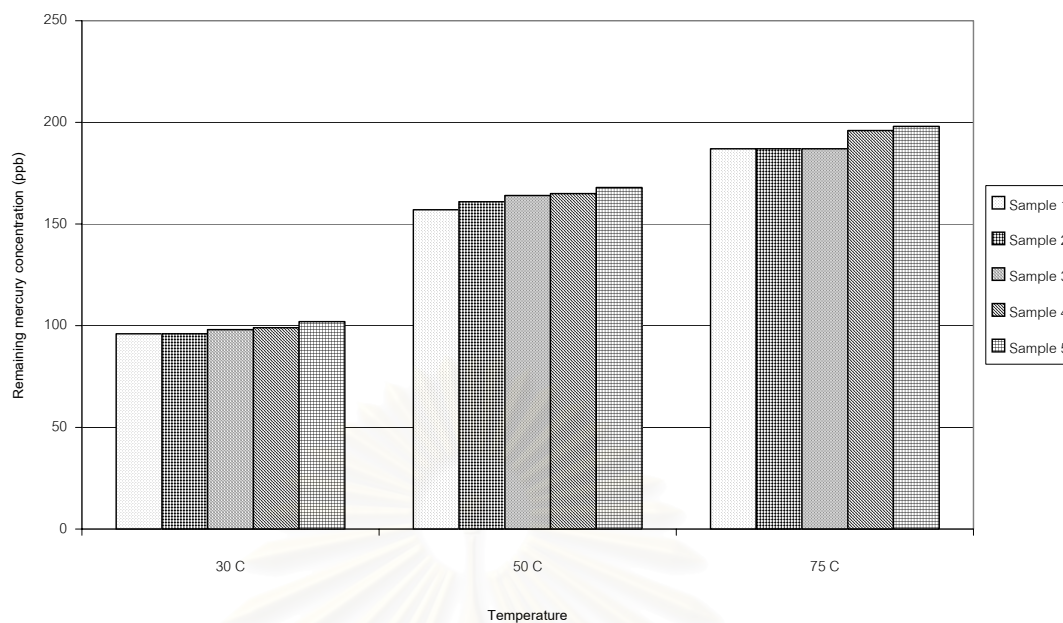


Figure 3.6 Remaining Mercury in Study on Error from Repeated Experiment of Mercuric Chloride at various Temperatures

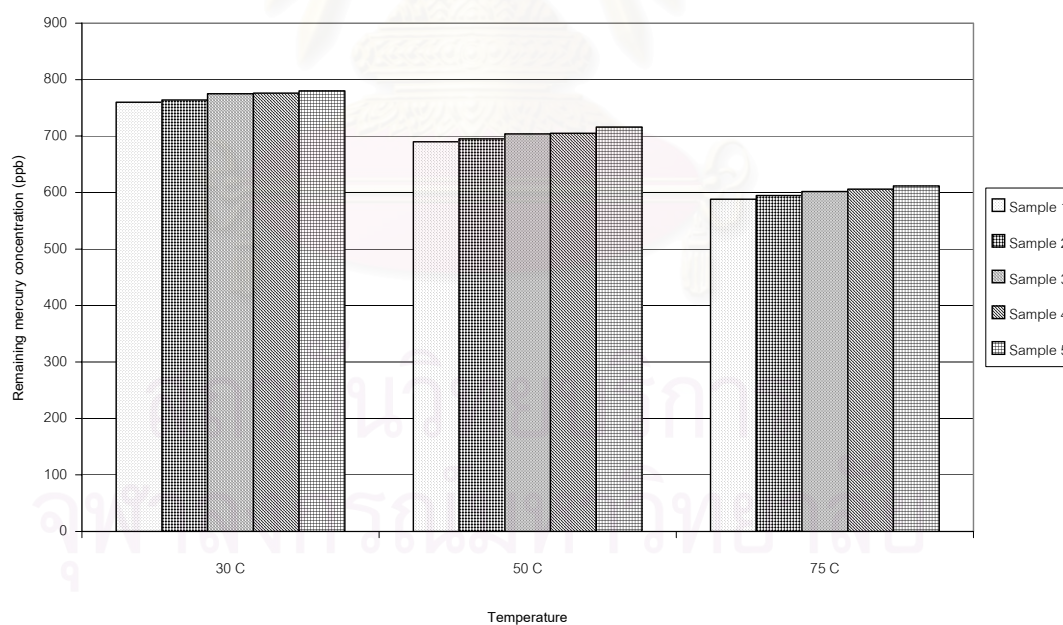


Figure 3.7 Remaining Mercury in Study on Error from Repeated Experiment of Diphenylmercury at various Temperatures

Table 3.2 Average Value of Concentration and Percent of Deviation Range of Mercury Compounds in repetition study.

Mercury Type	Initial conc. (ppb)	Temperature (°C)	Sample Con. (ppb)	Average Conc. (ppb)	% of Deviation Range
Mercuric Chloride	1000	30	96,96,98,99, 102	98.2	-2.24 to +3.87
		50	157,161,164, 165,168	163	-3.68 to +3.07
		75	187,187,187, 196,198	191	-2.09 to +3.66
Diphenylmercury	1000	30	760,764,775, 776,780	771	-1.43 to +1.17
		50	690,695,704, 705,716	702	-1.71 to +1.99
		75	588,595,602, 606,612	600.6	-2.10 to +1.90

From results above, it was certain that the value of remaining concentration for both of mercury compounds seem nearly constant. Table 3.2 shows percent of deviation range that is in range of 4% and 2% deviate from average concentration of mercuric chloride and diphenylmercury, respectively. That can consider to be neglected for both of mercury compounds.

Error from digestion

The experiments are conducted to study deviation from digestion error. The experiments are digestion of feedstock, both mercuric chloride and diphenylmercury, concentration about 1 ppm. The experiments are repeated at the same prepared-sample for 5 times. Experimental results are plotted in Figure 3.8. Deviation data are shown in Table 3.3.

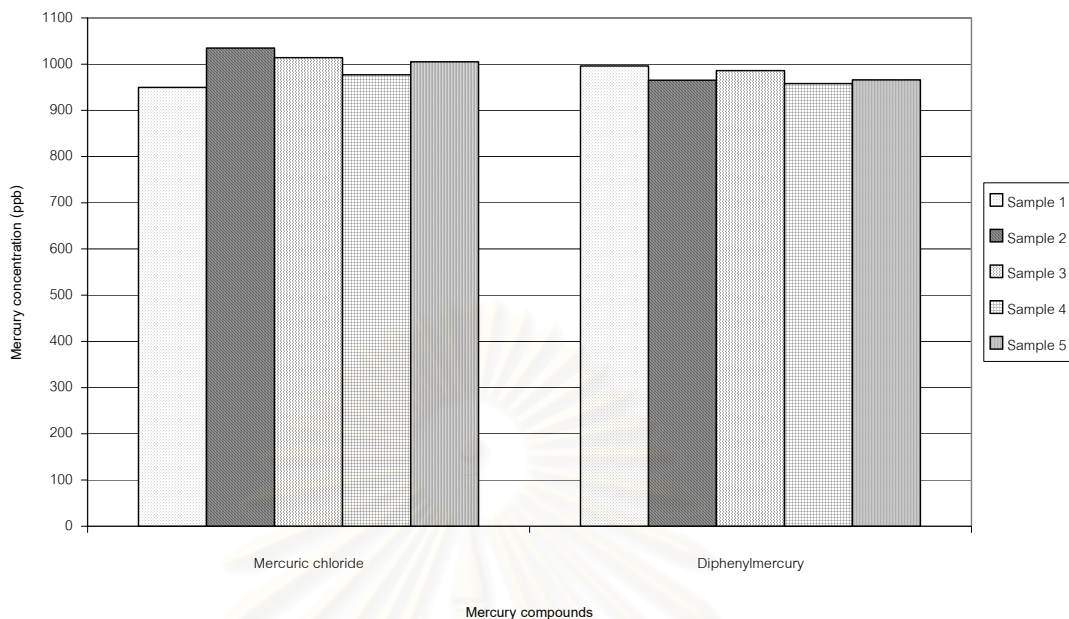


Figure 3.8 Concentration of Mercury Compounds in study on Error from digestion.

Table 3.3 Average Value of Concentration and Percent of Deviation Range of Mercury Compounds in digestion study.

Mercury Type	Sample Conc. (ppb)	Average Conc. (ppb)	% of Deviation Range
Mercuric Chloride	950, 1035, 1014, 977, 1005	996	-4.64 to +3.89
Diphenylmercury	996, 965, 986, 958, 966	974	-1.66 to +1.21

From results above, it was certain that the value of concentration after repeating digestion for both of mercury compounds seem nearly constant. Table 3.3 shows percent of deviation range that is in range of 5% and 2% deviate from average concentration of mercuric chloride and diphenylmercury, respectively. That can consider to be neglected the error from digestion for both of mercury compounds.

Error from Analysis by AAs

The studies are conducted to consider the deviation from analysis error. The experiments are detecting the samples, both mercuric chloride and diphenylmercury, concentration of samples before dilution and digestion about 1 ppm. The analyses are repeated at the same prepared-sample for 5 times. Experimental results are plotted in Figure 3.9. Deviation data are shown in Table 3.4.

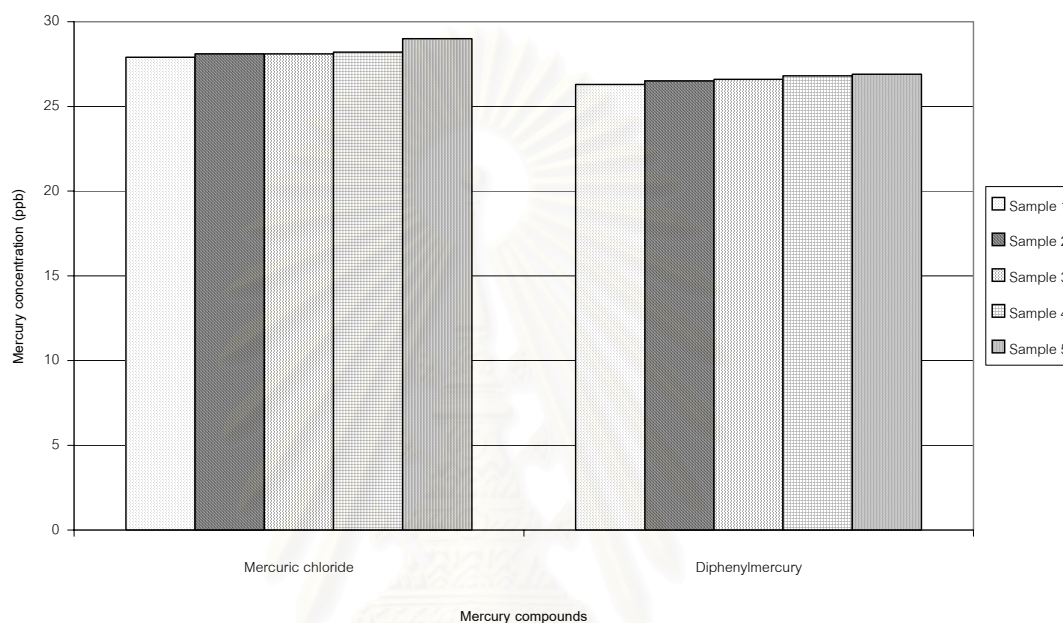


Figure 3.9 Samples concentration in study on Error from Analysis by AAs

Table 3.4 Average Value of Concentration and Percent of Deviation Range of Mercury Compounds in digestion study.

Mercury Type	Sample Conc. (ppb)	Average Conc. (ppb)	% of Deviation Range
Mercuric Chloride	27.9, 28.1, 28.1, 28.2, 29.0	28.3	-0.35 to +2.47
Diphenylmercury	26.3, 26.5, 26.6, 26.8, 26.9	26.6	-1.13 to +1.13

From results above, it certain that the value of concentration after repeating analysis for both of mercury compounds seem nearly constant. Table 3.4 shows percent of deviation range that is in range of 2.5% and 1.13% deviate from average concentration of mercuric chloride and diphenylmercury, respectively. The error can be from analysis by AAs considered to be neglected both of mercury compounds.

CHAPTER IV

RESULTS AND DISCUSSIONS

This chapter is divided into two parts: characteristics of adsorbents and experimental results.

Mercury removal experiments by adsorption were conducted in batch system. Mercuric chloride and diphenylmercury were used as model compounds for inorganic and organic mercury compounds, respectively. Toluene was used as a liquid hydrocarbon due to its high boiling point. After each experiment, liquid phase and solid phase were separated and analyzed for mercury content in each phase.

As shown in Chapter III, instrumental analysis, digestion and the experiment itself could cause errors in this study. It was found that the errors were less than 5%.

4.1 Characteristics of Adsorbents

There were five types of supports used in this study: alumina, silica, activated carbon, silica-alumina, and titanium oxide. Supplier, particle size, surface area, pore volume, average pore diameter and micropore area of these supports are summarized in Table 4.1.

Table 4.1 Supplier, Particle size, Total surface area, Pore volume, Average pore diameter and Micropore area of supports.

Support	Supplier	Particle size (micron)	Total surface area (m ² /g)	Pore volume (cc/g)	Average pore diameter (Å)	Micropore area (m ² /g)
Alumina	Aldrich	100	178.4	0.26	57.9	< 0.01
Silica	Carlo Erba	1.5	394.5	1.66	168.0	28
Activated carbon	Carbo-Karn	100	1120.2	0.64	18.1	822
Silica-alumina	Aldrich	100	545.2	0.80	58.4	< 0.01
Titanium oxide	Fluka	100	9.61	0.02	82.0	< 0.01

The average surface area is classified into 3 groups: micropore, mesopore and macropore. Table 4.2 shows the IUPAC classification of pores by size.

Table 4.2 Classification of pore sizes (Perry, R. H., 1998)

Type	Pore diameter, D (Å)
Micropore	D < 20
Mesopore	20 < D < 500
Macropore	500 < D

They showed that activated carbon is a micropore support. Average pore diameter of activated carbon is in the range of micropore. Micropore surface area of activated carbon is approximately 73.4% of total surface area.

Alumina, silica and silica-alumina are mesopore supports. Micropore surface area of silica is approximately 7% of total surface area. Alumina and silica-alumina supports have micropore surface area less than 0.01 m²/g.

Titanium oxide has low pore volume, approximately 8% of alumina and 1% of silica. It also has low surface area. These analyses indicate that titanium oxide may have no pore or its pore depth is low.

Each support was impregnated with copper nitrate solution and calcined in muffle furnace. Impregnation and calcination procedures are described in Chapter III.

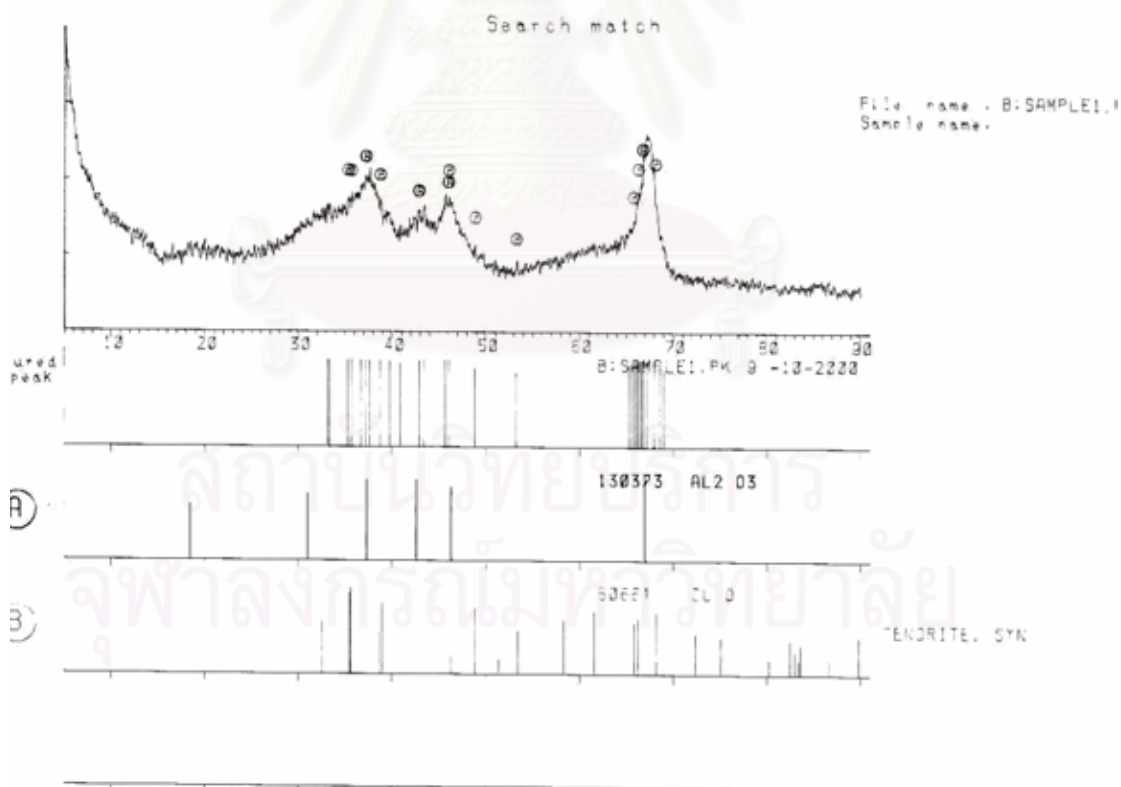
After impregnation and calcination, the adsorbents were analyzed for copper content using Flame Atomic Absorption Spectroscopy technique (AAs technique). The adsorbents were dissolved and decomposed before analysis. Details of procedure are described in Chapter III. The results of copper contents are listed in Table 4.3.

The range of copper content on support was from 2.20% to 2.34 % wt., which less than desired content of 2.5% wt.. The moisture content of copper nitrate used in this study measured by ASTM D-2216-98. The moisture content of copper nitrate used in this study was approximately 31%. Since copper nitrate was not dried before it was prepared for impregnation, its concentration in impregnation solution might not be the correct one. This error might be the cause of low copper content on the support.

Table 4.3 Copper content of copper adsorbent

Adsorbent	Copper content (% wt.)
CuO/alumina	2.34
CuO/silica	2.20
CuO/activated carbon	2.32
CuO/silica-alumina	2.25
CuO/titanium oxide	2.30

Chemical structures of copper oxide adsorbent were analyzed using X-Ray Diffraction (XRD) method. X-Ray diffraction patterns of copper oxide adsorbents are shown in Figure 4.1 to Figure 4.5. The results indicated that copper was in the form of copper oxide (CuO). Surface area, pore volume, average pore diameter and micropore area of copper adsorbents were analyzed by BET method. The results are shown in Table 4.4.

**Figure 4.1** XRD pattern of CuO/alumina

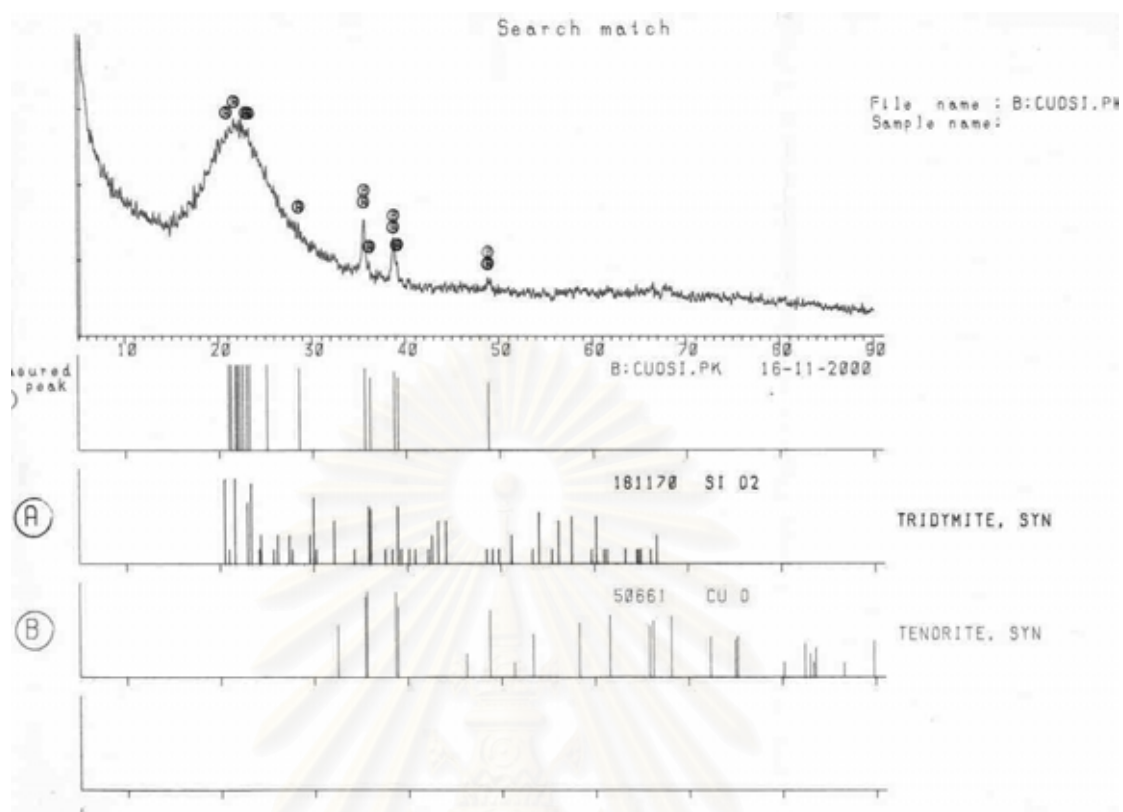


Figure 4.2 XRD pattern of CuO/silica

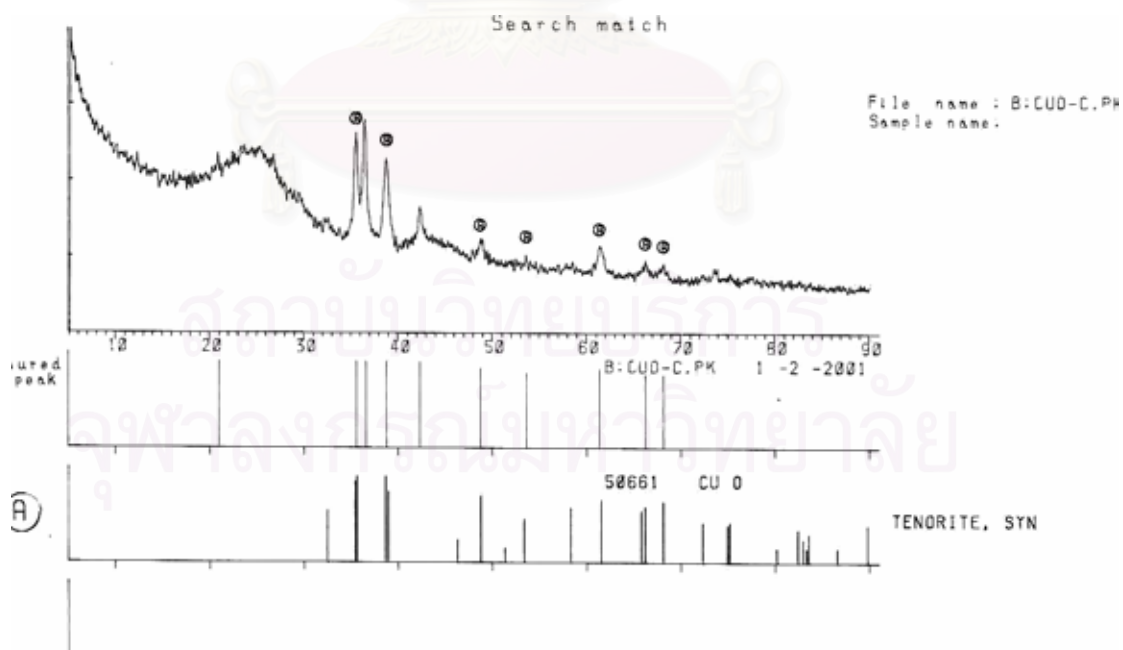


Figure 4.3 XRD pattern of CuO/activated carbon

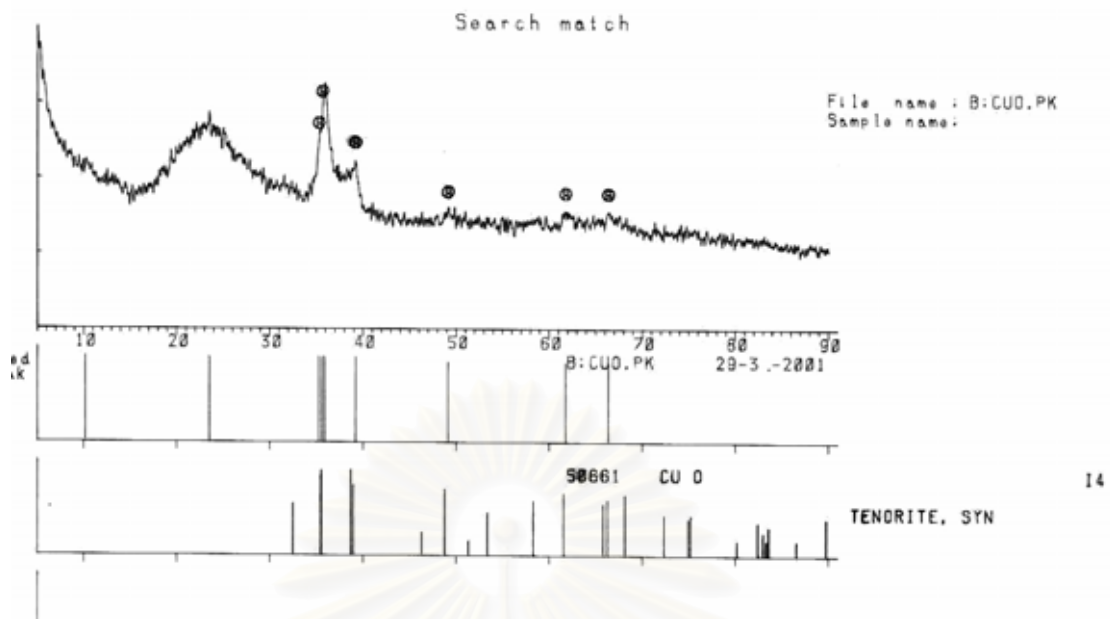


Figure 4.4 XRD pattern of CuO/silica-alumina

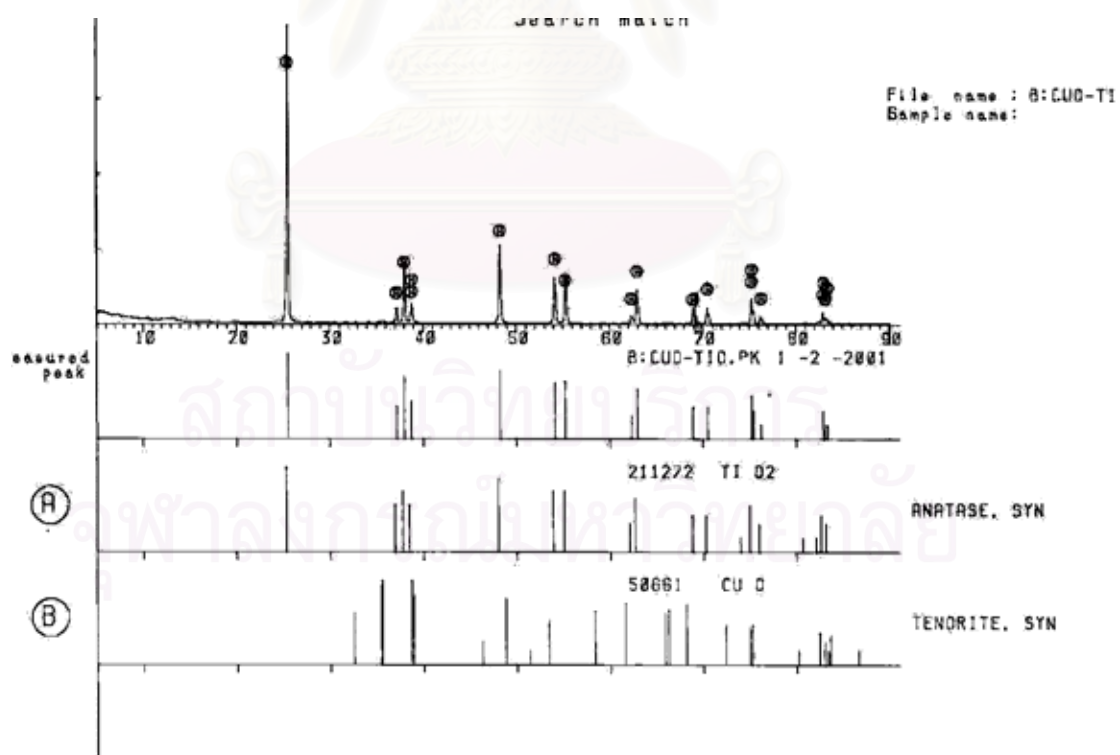


Figure 4.5 XRD pattern of CuO/titanium oxide

Table 4.4 Surface area, pore volume, average pore diameter and micropore area of adsorbents

Adsorbent	Total surface area (m ² /g)	Pore volume (cc/g)	Average pore diameter (Å)	Micropore surface area (m ² /g)
Alumina	178.4	0.26	57.9	< 0.01
CuO/alumina	149.4	0.21	52.3	<0.01
Silica	394.5	1.66	168.0	28
CuO/silica	343.3	1.58	154.0	22
Activated carbon	1120.2	0.64	18.1	822
CuO/activated carbon	966.8	0.59	16.0	801
Silica-alumina	545.2	0.80	58.4	< 0.01
CuO/silica-alumina	516.3	0.72	55.4	< 0.01
Titanium oxide	9.61	0.02	82.0	< 0.01
CuO/titanium oxide	12.1	0.037	121.5	< 0.01

It is clearly shown that surface area, pore volume and average pore diameter of each type of copper oxide adsorbents, except CuO/TiO₂, decreased slightly when they were compared with the supports. Surface area, pore volume and average pore diameter decreased approximately 5-15%, 5-20% and 5-12%, respectively.

Micropore surface area of CuO/activated carbon and CuO/silica decreased slightly when they are compared with the supports.

The decrement of surface area of copper oxide adsorbents, except CuO/TiO₂, may be resulted from the decrement of average pore diameter or the pore is blocked by copper oxide. To verify this assumption, pore structure of support was assumed to be cylindrical pore and the surface area of adsorbent was internal area of the cylinder. The pore depth could be calculated by Equation 4.1. The results of pore depth calculated are shown in Table 4.5.

$$\text{Pore depth} = \frac{\text{Surface area}}{\pi D} ; D = \text{average pore diameter} \dots\dots\dots(4.1)$$

Table 4.5 Calculated pore depth of adsorbent

Adsorbent	Pore depth (m/g) *10 ⁻¹⁰	Change
Alumina	0.98	-7%
CuO/alumina	0.91	
Silica	0.75	-5%
CuO/silica	0.71	
Activated carbon	19.7	-3%
CuO/activated carbon	19.2	
Silica-alumina	2.97	-0.3%
CuO/silica-alumina	2.96	

A slight change in pore depth indicates that copper oxide, impregnated on the support, do not block any part of the pore. The decrement of surface area was resulted from the decrement of average pore diameter.

4.2 Experimental results and discussions

Table 4.6 shows the operating conditions of experiment. After each experiment, solid phase (adsorbent) and liquid phase were separated. Mercury concentration in liquid phase and solid phase were analyzed using digestion and AAs technique, which were described in Chapter III. The results of all experiment are shown in Appendix A.

Table 4.6 Operating conditions of experiment

Mercury compounds	1. Mercuric chloride (HgCl ₂) 2. Diphenylmercury (C ₁₂ H ₁₀ Hg)
Solvent	Toluene
Initial concentration	1000 ppb (adsorption), 0 ppb (desorption)
Dose of each experiment	1 g of adsorbent : 100 g of toluene
Temperature	1. 30°C 2. 50°C 3. 75°C
Adsorbent	1. Alumina 2. Silica 3. Activated carbon 4. Silica-alumina 5. Titanium oxide 6. CuO/alumina 7. CuO/silica 8. CuO/activated carbon 9. CuO/silica-alumina 10. CuO/titanium oxide

In each experiment, mercury concentration of the feed was analyzed and it was found that mercury concentration was not equal to exactly 1000 ppb. The range of mercury concentration of feed was from 950 ppb to 1050 ppb. The percent removal was defined to compare adsorptive ability of each adsorbent. The amount of adsorbed mercury was calculated by Equation 4.2. The percent removal was calculated by Equation 4.3. The adsorptive ability was also depending on the surface area of adsorbent. The amount of adsorbed mercury per surface area was calculated (Equation 4.4) to determine the adsorptive ability of supports and adsorbents.

Amount of adsorbed mercury (μg)

$$= \frac{\text{mercury conc. of feed (ppb)} - \text{mercury conc. of effluent (ppb)}}{\dots\dots\dots}(4.2)$$

$$\text{Percent removal} = \frac{\text{amount of adsorbed mercury} \times 1000}{\text{Mercury concentration of feed (ppb)}} \dots\dots\dots(4.3)$$

$$\begin{aligned} &\text{Amount of adsorbed mercury per surface area } (\mu\text{g/g}) \\ &= \frac{\text{amount of adsorbed mercury}}{\text{total surface area of adsorbent or support}} \dots\dots\dots(4.4) \end{aligned}$$

Some of spent supports and copper oxide adsorbents were classified and used in XRD analysis to detect crystalline structure substance on their surface, used in analysis of the amount of mercury that deposited on the surface, and used in the study of mercury desorption. The results of the study of desorption are reported in percent desorption that can be calculated by Equation 4.5.

$$\text{Percent desorption} = \frac{\text{amount of desorbed mercury} \times 100}{\text{amount of adsorbed mercury}} \dots\dots\dots(4.5)$$

4.2.1 Adsorption of mercury compounds by supports

Table 4.7 shows percent removal of mercuric chloride and amount of adsorbed mercury per surface area by supports.

The results showed that alumina, activated carbon had high percent removal. Percent removal of alumina and activated carbon were more than 50% at all operating temperature. The ranges of percent removal of silica-alumina and titanium oxide were from 20% to 60% (at 75°C). In case of silica, it had low percent removal (<20%) at all operating temperature.

On the contrary, amount of adsorbed mercury per surface of titanium oxide was very high. It indicated that surface of titanium oxide was the most effective surface. In case of activated carbon, which had high percent removal, its surface was less effective than titanium oxide and alumina. It indicated that percent removal of activated carbon was high due to it had a lot of surface area.

Table 4.7 Removal of mercuric chloride in study of adsorption on support at various temperatures.

Adsorbent	Temperature (°C)	Percent removal	Adsorbed mercury per surface area ($\mu\text{g}/\text{m}^2$)
Alumina	30	82.4	0.46
	50	70.2	0.39
	75	53.4	0.30
Silica	30	16.8	0.043
	50	18.1	0.046
	75	20.1	0.051
Activated carbon	30	76.0	0.068
	50	75.5	0.067
	75	76.0	0.068
Silica-alumina	30	16.5	0.030
	50	25.0	0.046
	75	39.4	0.072
Titanium oxide	30	29.8	3.10
	50	43.0	4.47
	75	59.8	6.22

The results were rearranged to show the effect of temperature on mercuric chloride removal in Figure 4.6. Figure 4.6 shows percent removal of each support versus operating temperature. The results showed that operating temperature had effect on removal of mercuric chloride of some supports. Percent removal of mercuric chloride decreased with increasing operating temperature for alumina support. Percent removal of mercuric chloride appeared constant with operating temperature for silica and activated carbon support. Percent removal of mercuric chloride increased with increasing operating temperature for silica-alumina and titanium oxide supports.

The results indicated that the adsorption of mercuric chloride on alumina supports was physical adsorption.

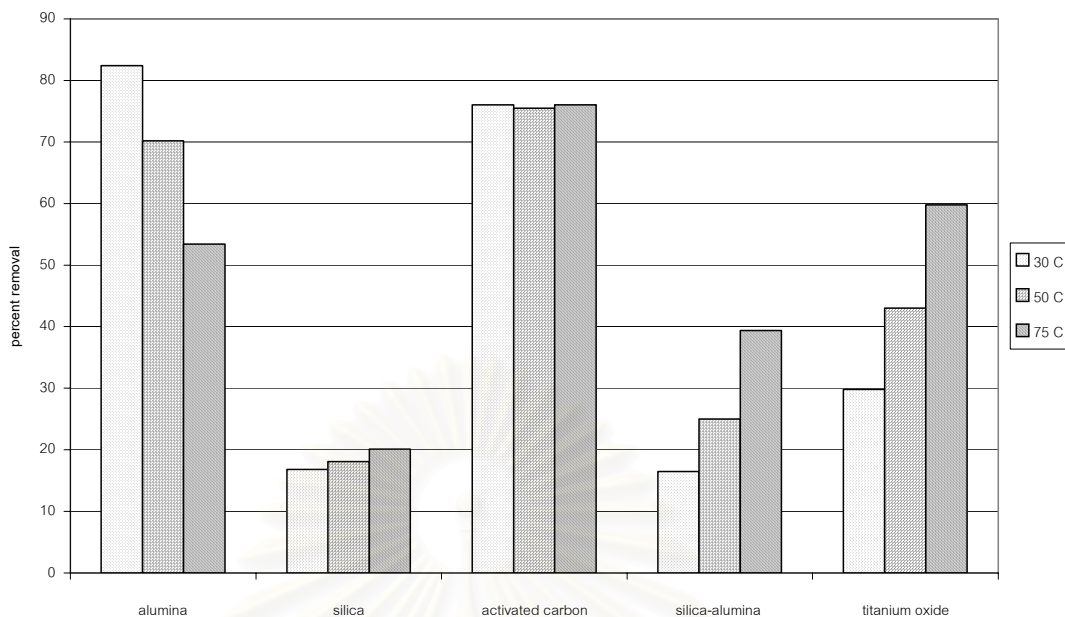


Figure 4.6 Percent of mercuric chloride removed in the study of adsorption by supports at various temperatures.

The XRD patterns of spent support did not show the presence of crystalline substance on surface of supports. In case of alumina support, Sookkho (1995) and Chokelarb (2000) also obtained similar result. Their XRD patterns were not found crystalline substance on surface of alumina in the study of mercuric chloride adsorption.

The results of support digestion and desorption are shown in Table 4.8. The results showed that the amount of mercuric chloride digested from support was approximately 75-80% of the amount of adsorbed mercury. The error may occur from adsorbent digestion.

The desorption study showed that mercuric chloride could desorb from the spent supports. The percent desorption of alumina, silica, silica-alumina and titanium oxide were approximately 20-22%. The percent desorption of activated carbon was approximately 12%.

Table 4.8 Amount of digested mercuric chloride and desorbed mercuric chloride from spent supports

Support	Adsorbed mercury at 75°C (µg/g)	Digested mercury (µg/g)	Desorbed mercury (µg/g)	Percent desorption
Alumina	53.4	40.1	11.5	21.5
Silica	20.1	16.1	4.1	20.3
Activated carbon	76.0	59.0	9.2	12.1
Silica-alumina	39.4	31.1	8.7	22.0
Titanium oxide	59.8	49.8	13.0	21.7

Table 4.9 shows percent removal and amount of adsorbed mercury per surface area in the study of diphenylmercury adsorption by supports. The results showed that the effective supports for adsorb diphenylmercury were alumina and activated carbon. The other supports had low percent removal for diphenylmercury. On the contrary, titanium oxide and alumina had high amount of adsorbed mercury per surface area. The other supports had low amount of adsorbed mercury per surface area.

The results were rearranged to show the effect of temperature on diphenylmercury removal in Figure 4.7. Figure 4.7 shows percent removal of each support versus operating temperature. The results showed that operating temperature had effect on removal of diphenylmercury of some supports. Percent removal of diphenylmercury increased with increasing operating temperature for alumina and activated carbon support. Percent removal of diphenylmercury appeared constant with operating temperature for silica, silica-alumina and titanium oxide supports.

The XRD patterns of spent supports did not show the presence of crystalline substance on surface of support. In case of alumina support, Sookkho (1995) and Chokelarb (2000) also obtained similar result. Their XRD patterns were not found crystalline substance on surface of alumina in the study of diphenylmercury adsorption.

Table 4.9 Removal of diphenylmercury in study of adsorption on support at various temperatures.

Adsorbent	Temperature (°C)	Percent removal	Adsorbed mercury per surface area ($\mu\text{g}/\text{m}^2$)
Alumina	30	9.5	0.053
	50	16.2	0.091
	75	28.4	0.160
Silica	30	3.1	0.008
	50	4.0	0.010
	75	5.9	0.015
Activated carbon	30	15.0	0.013
	50	17.1	0.015
	75	24.9	0.022
Silica-alumina	30	6.1	0.011
	50	7.2	0.013
	75	9.4	0.017
Titanium oxide	30	2.7	0.28
	50	3.5	0.36
	75	3.7	0.39

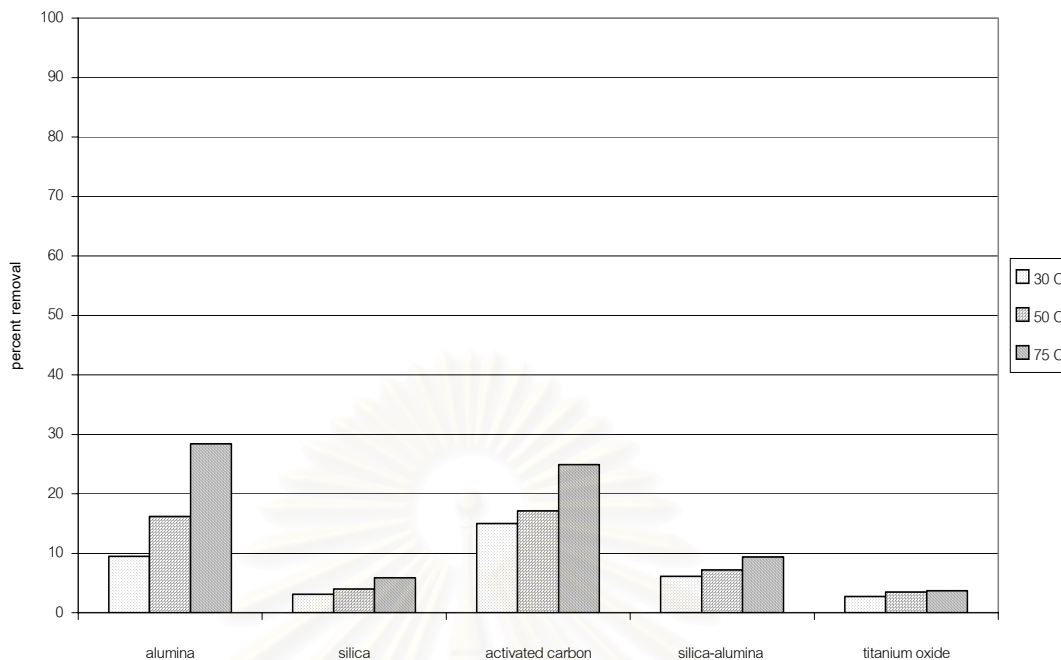


Figure 4.7 Percent removal of diphenylmercury in the study of adsorption by supports at various temperatures.

Table 4.10 shows the amount of adsorbed mercury, amount of mercury that digested from supports, amount of mercury that desorbed from supports, and percent desorption in the study of adsorption of diphenylmercury by supports. The results showed that the amount of diphenylmercury digested from support is approximately 67-85% of the amount of adsorbed mercury.

The results also showed that diphenylmercury could desorb from the spent supports. The percent desorption of alumina, silica, silica-alumina and titanium oxide were approximately 23-25%. The percent desorption of activated carbon was approximately 16.2%.

Table 4.10 Amount of digested diphenylmercury and desorbed diphenylmercury from spent supports

Support	Adsorbed mercury at 75°C (µg/g)	Digested mercury(µg/g)	Desorbed mercury (µg/g)	Percent desorption
Alumina	28.4	24.1	6.5	23.0
Silica	5.9	4.4	1.42	24.1
Activated carbon	24.9	20.0	4.0	16.2
Silica-alumina	9.4	7.0	2.2	23.6
Titanium oxide	3.7	2.5	0.9	25.1

The results also showed that mercuric chloride could be adsorbed by support with higher adsorptive ability than diphenylmercury. It indicates that type of mercury compound strongly affect the adsorption of mercury on supports. Mercuric chloride has high polarities while diphenylmercury has more complicated. Sookkho (1995) suggested that polarity of mercury compounds is one of properties, which can effect on interaction between adsorbent and mercury. He found that percent removal of mercury increased with increasing of polarity of the compounds. Soontaranurak (1998) suggested that mercuric chloride which has high polarity can adsorb on the surface of chromium oxide film by sharing electron. Diphenylmercury has the strongest metal-carbon bond of the common organic mercury compound. Diphenylmercury have two aromatic rings, which are stable. In this study, diphenylmercury that is classified as organic mercury compounds can be partially removed by adsorption on supports. On a contrary, Yamada (1995), who studied adsorption of organic mercury compound suggested that mercury compounds, especially organic mercury compounds, could not be adsorbed on any types of adsorbents but it could be decomposed and converted into elemental mercury, then it is adsorbed on suitable adsorbents. In his study, the decomposition of mercury compounds was conducted at temperatures of 165°C to 300°C. In this study, adsorption experiments were conducted at temperatures less than 80°C in which diphenylmercury were not expected to decompose. Structure of mercury compound adsorbed on the surface of the supports can not be identified, it is believed that diphenylmercury in this study does not decompose upon adsorption but it adsorbs directly on to support surface.

Mercuric chloride, which is classified as ionic mercury compounds, can be adsorbed on support. Remy (1956) suggested that mercuric chloride is different from other metal chloride. Metal chloride, when dissolves into water, it decomposes to metal ions and chloride ions but does not mercuric chloride. This is accord with other authors such as Biscarini (1971) and Gomez (1997). Thus, mercuric chloride is directly adsorb on alumina surface.

4.2.2 Adsorption of mercury compounds by 2.5 % copper oxide adsorbents

Table 4.11 shows percent removal of mercuric chloride and amount of adsorbed mercury per surface area of copper oxide adsorbents. CuO/titanium oxide had highest amount of adsorbed mercury per surface area and CuO/activated carbon had high percent removal but low amount of adsorbed mercury per surface area. The similar results were obtained from the study of supports.

The results were rearranged to show the effect of temperature on mercuric chloride removal in Figure 4.8. Figure 4.8 shows percent removal of each adsorbent versus operating temperature. The results showed that operating temperature had effect on percent removal of mercuric chloride of some adsorbents. Percent removal of mercuric chloride decreased with increasing operating temperature for CuO/alumina adsorbent. Percent removal of mercuric chloride appeared constant with operating temperature for CuO/ activated carbon adsorbent. Percent removal of mercuric chloride increased with increasing operating temperature for CuO/silica, CuO/silica-alumina and CuO/titanium oxide adsorbents. The same trend was also observed in the study of their support.

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Table 4.11 Removal of mercuric chloride in study of adsorption on 2.5 % copper adsorbent at various temperatures.

Adsorbent	Temperature (°C)	Percent removal	Adsorbed mercury per surface area ($\mu\text{g}/\text{m}^2$)
CuO/Alumina	30	90.2	0.60
	50	83.5	0.56
	75	79.7	0.53
CuO/Silica	30	24.5	0.071
	50	30.3	0.088
	75	36.4	0.106
CuO/Activated carbon	30	88.8	0.092
	50	87.4	0.090
	75	86.3	0.089
CuO/Silica-alumina	30	28.6	0.055
	50	46.8	0.091
	75	58.3	0.113
CuO/Titanium oxide	30	45.6	3.769
	50	53.4	4.413
	75	74.6	6.165

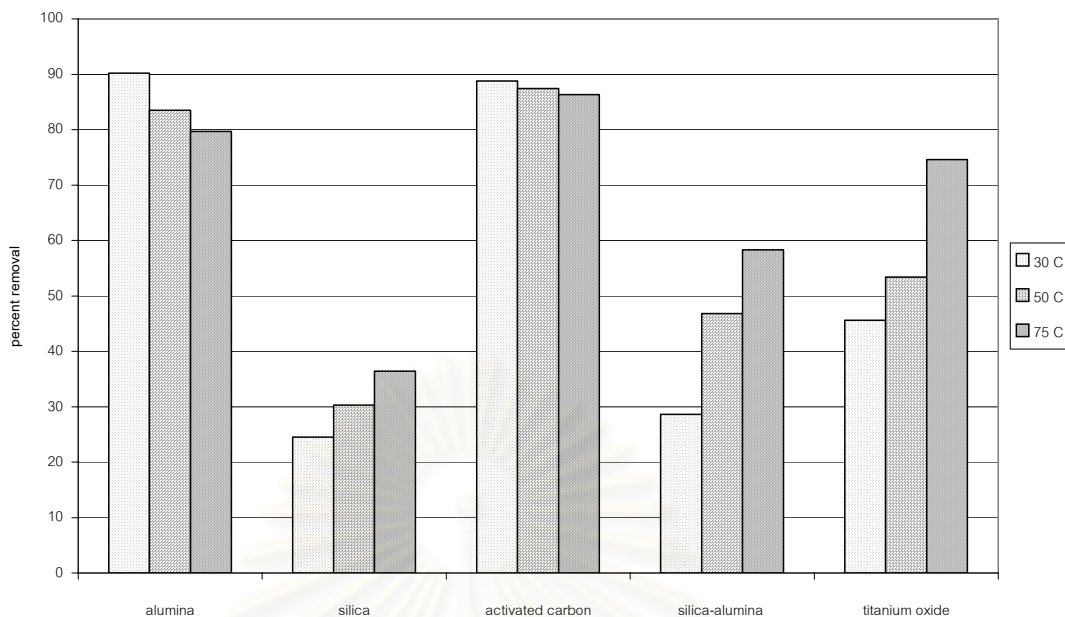


Figure 4.8 Percent removal of mercuric chloride in the study of adsorption by 2.5 % Copper oxide adsorbents at various temperatures.

Some of spent copper oxide adsorbents were analyzed by XRD method to find crystalline substance on adsorbents surface. CuO/alumina, CuO/silica-alumina and CuO/titanium oxide were selected to analyze. From XRD analysis, the XRD patterns of spent adsorbent showed the presence of chemical bond between mercury and copper (CuHg and $\text{Cu}_{15}\text{Hg}_{11}$). Chokelarb (2000), who studied adsorption of mercury by CuO/alumina, also obtained similar result. Figure 4.9 to Figure 4.11 show XRD pattern of copper mercury.

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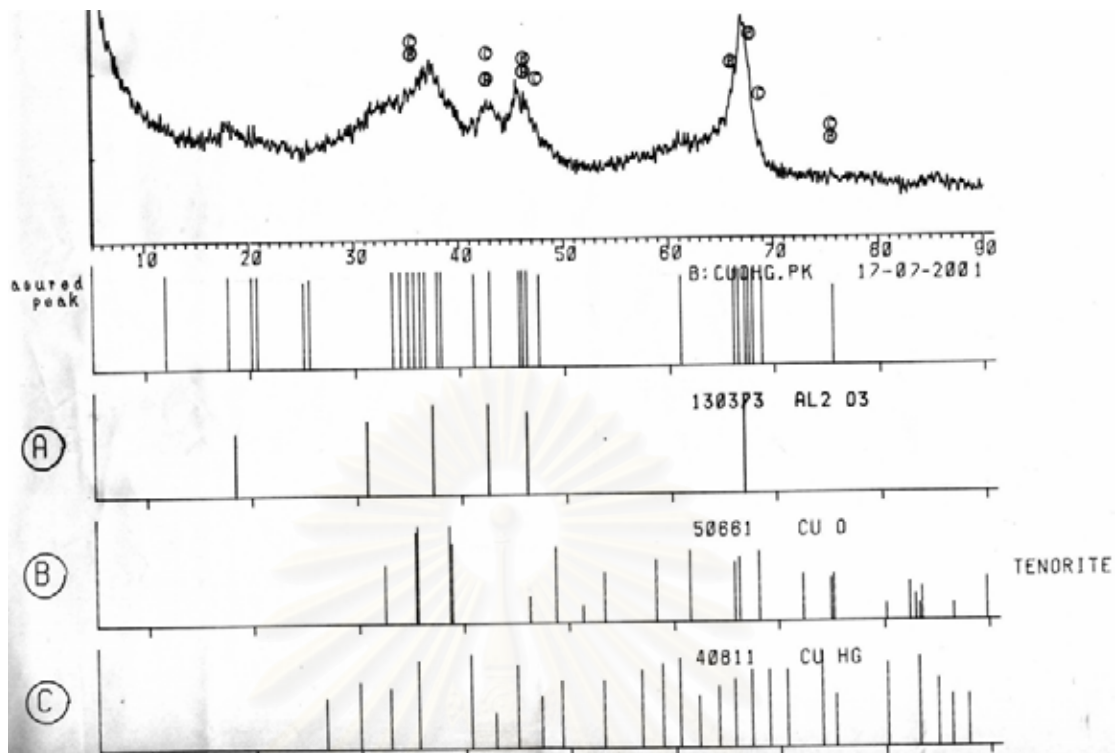


Figure 4.9 XRD pattern of spent 2.5 % CuO/alumina adsorbent for mercuric chloride

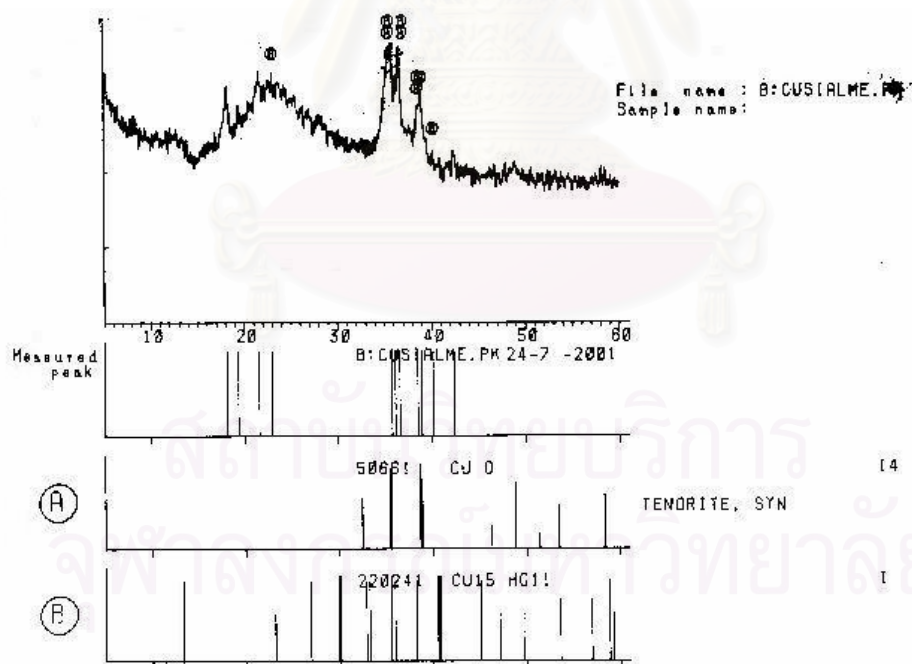


Figure 4.10 XRD pattern of spent 2.5 % CuO/silica-alumina adsorbent for mercuric chloride.

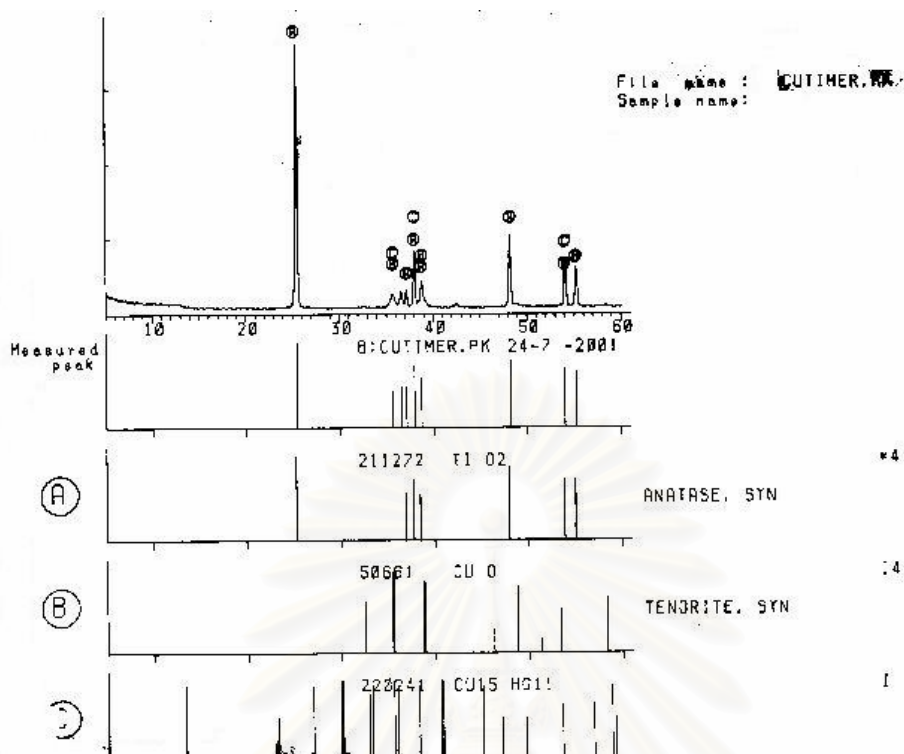


Figure 4.11 XRD pattern of spent 2.5 % CuO/titanium oxide adsorbent for mercuric chloride.

The results of adsorbents digestion and desorption are shown in Table 4.12. The results showed that the amount of mercuric chloride digested from adsorbents were approximately 75-80% of the amount of adsorbed mercury.

The percent desorption of alumina, silica, silica-alumina and titanium oxide are also show in Table 4.11 to compare with copper oxide adsorbent. The percent desorption of CuO/alumina, CuO/silica, CuO/silica-alumina and CuO/titanium oxide were approximately 13-15%. The percent desorption of CuO/activated carbon was approximately 7.5%.

The results showed that the amounts of mercuric chloride adsorbed by copper oxide adsorbents were more than the amounts of mercuric chloride adsorbed by supports but the amounts of mercuric chloride desorbed from copper oxide adsorbents were less than the amounts of mercuric chloride desorbed from supports. The amount of mercuric chloride desorbed from copper oxide adsorbent was closed to amount of mercuric chloride desorbed from each support.

Table 4.12 Amount of digested mercuric chloride and desorbed mercuric chloride from spent adsorbents

Support	Adsorbed mercury at 75°C (µg/g)	Digested mercury (µg/g)	Desorbed mercury (µg/g)	Percent desorption
Alumina	53.4	40.1	11.5	21.5
Silica	20.1	16.1	4.1	20.3
Activated carbon	76.0	57.8	9.2	12.1
Silica-alumina	39.4	31.5	8.7	22.0
Titanium oxide	59.8	47.8	13.0	21.7
CuO/alumina	79.7	59.8	12.3	15.4
CuO/silica	36.4	28.4	5.3	14.5
CuO/activated carbon	86.3	69.0	6.5	7.5
CuO/silica-alumina	58.3	44.3	7.6	13.0
CuO/titanium oxide	74.6	59.7	10.3	13.8

Table 4.13 shows percent removal of diphenylmercury by copper oxide adsorbents. The amounts of adsorbed mercury per surface area of adsorbent also show in Table 4.13. The results showed that percent removal of mercuric chloride increased approximately 5-10% from percent removal of supports. CuO/titanium oxide had highest amount of mercury adsorb per surface area and CuO/activated carbon had high percent removal but low amount of mercury per surface area. The similar results were obtained from the study of supports.

Table 4.13 Removal of diphenylmercury in study of adsorption on 2.5 % copper adsorbent at various temperatures.

Adsorbent	Temperature (°C)	Percent removal	Adsorbed mercury per surface area ($\mu\text{g}/\text{m}^2$)
CuO/Alumina	30	19.9	0.13
	50	25.4	0.17
	75	37.6	0.25
CuO/Silica	30	8.0	0.023
	50	9.5	0.028
	75	12.5	0.036
CuO/Activated carbon	30	29.4	0.030
	50	28.3	0.029
	75	31.2	0.032
CuO/Silica-alumina	30	13.3	0.026
	50	17.7	0.034
	75	20.4	0.040
CuO/Titanium oxide	30	6.1	0.504
	50	8.5	0.702
	75	12.0	0.992

The results were rearranged to show the effect of temperature on diphenylmercury removal in Figure 4.12. Figure 4.12 shows percent removal of each adsorbent versus operating temperature. The results showed that operating temperature has effect on percent removal of diphenylmercury of some adsorbents. Percent removal of diphenylmercury increased with increasing operating temperature for CuO/alumina adsorbent. Percent removal of diphenylmercury appeared nearly constant with operating temperature for CuO/silica, CuO/activated carbon, CuO/silica-alumina and CuO/titanium oxide adsorbent.

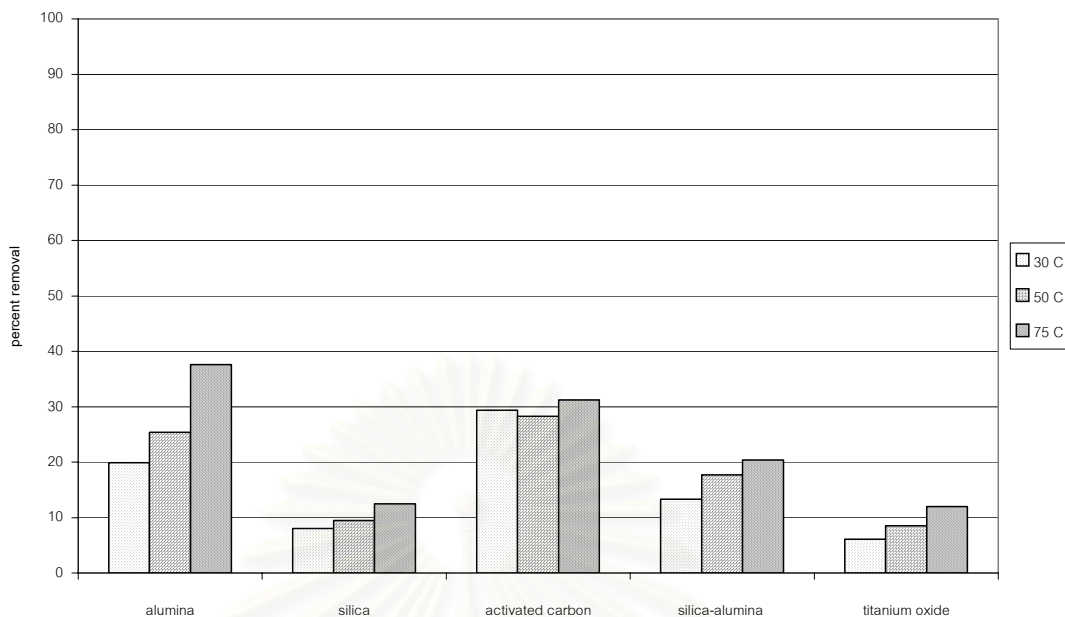


Figure 4.12 Percent removal of diphenylmercury in the study of adsorption by 2.5 % Copper oxide adsorbents at various temperatures.

Some of spent copper oxide adsorbents were analyzed by XRD method to find crystalline substance on adsorbents surface. CuO/alumina, CuO/silica and CuO/activated carbon were selected to analyze. The XRD patterns of spent adsorbents showed the presence of chemical bond between mercury and copper (CuHg). Chokelard (2000), who studied adsorption of diphenylmercury by CuO/alumina, also found chemical bond between mercury and copper (CuHg). Figure 4.13 to Figure 4.15 show XRD pattern of CuHg.

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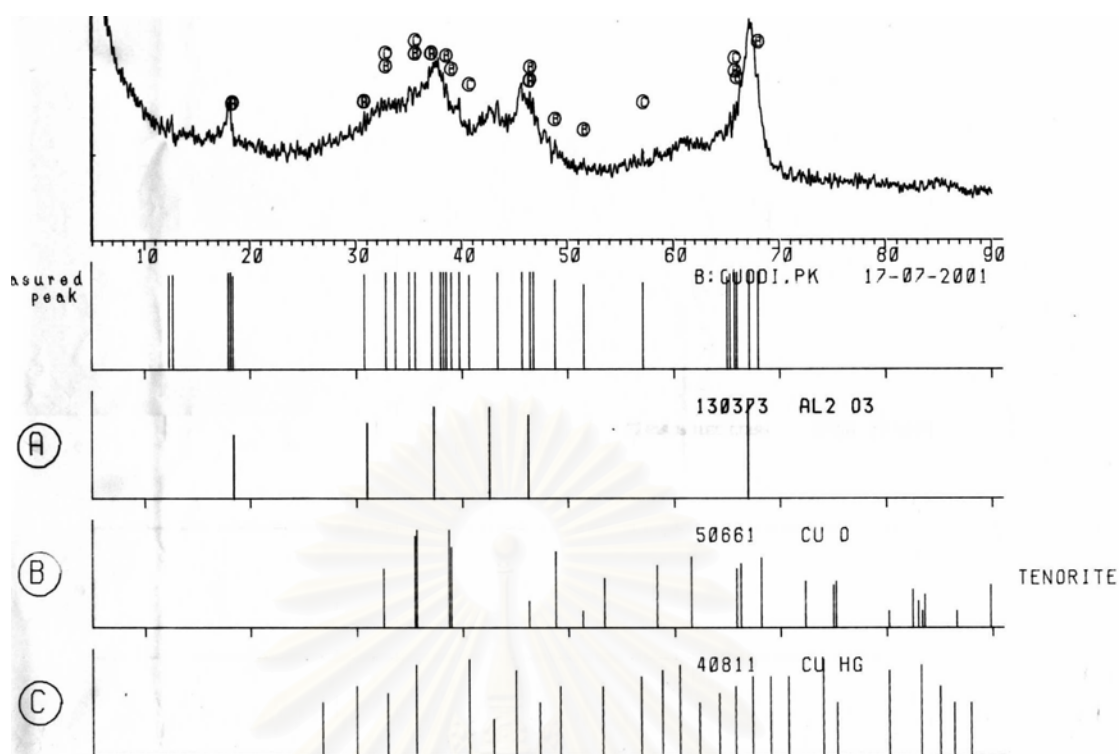


Figure 4.13 XRD pattern of spent 2.5 % CuO/alumina adsorbent for diphenylmercury

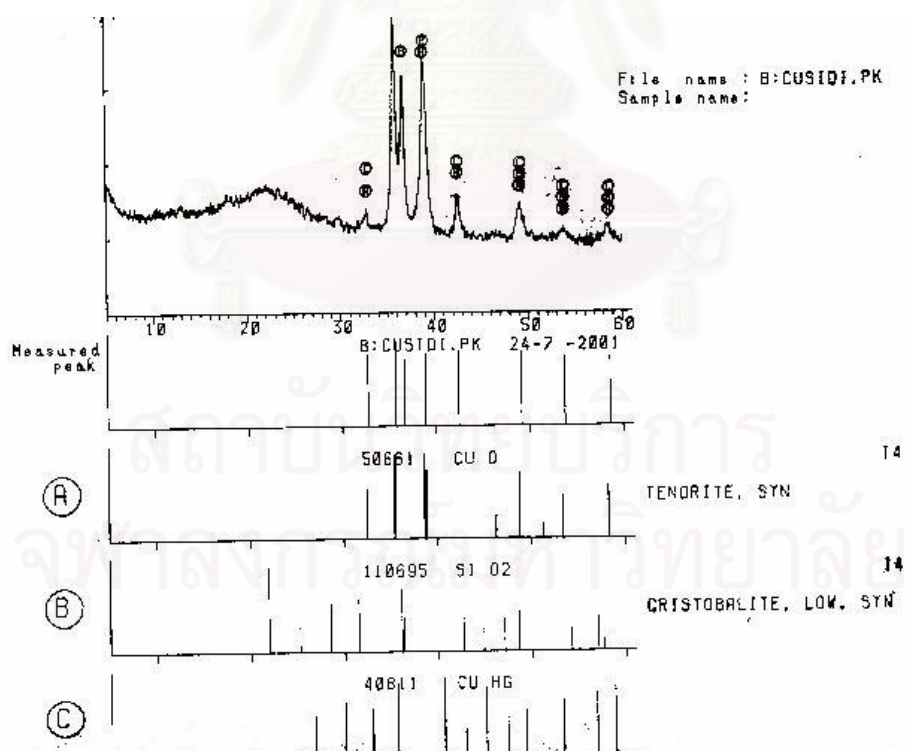


Figure 4.14 XRD pattern of spent 2.5 % CuO/silica adsorbent for diphenylmercury

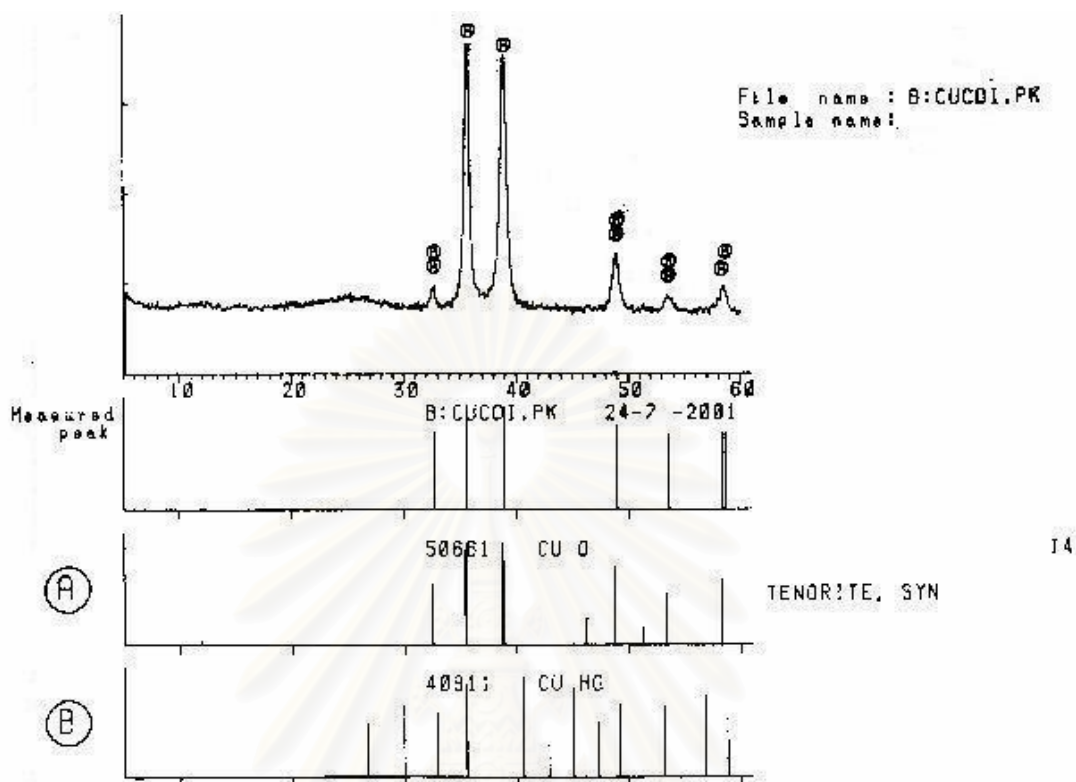


Figure 4.15 XRD pattern of spent 2.5 % CuO/activated carbon adsorbent for diphenylmercury

The amount of mercury digested from adsorbents is shown in Table 4.14. The results showed that the amount of diphenylmercury digested from adsorbents was approximately 75-80% of the amount that adsorbed by adsorbents.

The amount of mercury desorbed from spent copper oxide adsorbents is also shown in Table 4.14. The percent desorption of alumina, silica, silica-alumina and titanium oxide are also show in Table 4.14 to compare with copper oxide adsorbent. The percent desorption of CuO/alumina, CuO/silica, CuO/silica-alumina and CuO/titanium oxide were approximately 16-18%. The percent desorption of CuO/activated carbon was approximately 10%.

The results showed that the amounts of diphenylmercury adsorbed by copper oxide adsorbents were more than the amounts of diphenylmercury adsorbed by supports but the amounts of diphenylmercury desorbed from copper oxide adsorbents

were less than the amounts of diphenylmercury desorbed from supports. The amount of diphenylmercury desorbed from copper oxide adsorbent was closed to amount that desorbed from each support.

Table 4.14 Amount of digested diphenylmercury and desorbed diphenylmercury from spent adsorbents

Support	Adsorbed mercury at 75°C (µg/g)	Digested mercury (µg/g)	Desorbed mercury (µg/g)	Percent desorption
Alumina	28.4	24.14	6.5	23.0
Silica	5.9	5.0	1.42	24.1
Activated carbon	24.9	20.0	4.0	16.2
Silica-alumina	9.4	7.0	2.2	23.6
Titanium oxide	3.7	2.8	0.9	25.1
CuO/alumina	37.6	32.0	6.0	16.0
CuO/silica	12.5	9.5	2.0	15.4
CuO/activated carbon	31.2	25.0	3.3	10.6
CuO/silica-alumina	20.4	16.0	3.4	16.7
CuO/titanium oxide	12.0	10.1	2.2	18.0

The results showed that mercuric chloride could be removed more effectively than diphenylmercury by all copper oxide adsorbents. The similar results were obtained from the study of supports. This indicated that type of mercury compound strongly affect the adsorption of mercury on copper oxide adsorbents. Sookkho (1995) suggested that polarity of mercury compounds is one of properties, which can effect in interaction between adsorbent and mercury. He found that percent removal of mercury increased with increasing of polarity of compounds.

CHAPTER V

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

The following conclusions are drawn from this study

1. Alumina, silica, activated carbon, silica-alumina and titanium oxide supports can adsorb mercuric chloride and diphenylmercury in liquid hydrocarbon.
2. Impregnation of copper oxide on the supports increases amount of mercuric chloride and diphenylmercury adsorbed on the adsorbents.
3. Percent removal of mercuric chloride and diphenylmercury depend on type of support. In case of mercuric chloride, the percent removal of adsorbent is in following order: CuO/activated carbon > CuO/alumina > CuO/titanium oxide > CuO/silica-alumina > CuO/silica. In case of diphenylmercury, the percent removal of adsorbent is in following order: CuO/activated carbon > CuO/alumina > CuO/silica-alumina > CuO/titanium oxide > CuO/silica.
4. Percent removal of mercuric chloride and diphenylmercury depend on operating temperature. In case of diphenylmercury, the percent removal increases with increasing operating temperature. In case of mercuric chloride, the percent removal of alumina and CuO/alumina decrease with increasing operating temperature. The percent removal of other adsorbents increase with increasing operating temperature.
5. Percent removal of mercury compounds depend on type of mercury. Mercuric chloride can be removed more effectively than diphenylmercury by all supports and adsorbents.

Recommendations

1. A similar study should be conducted in continuous process such as fixed bed adsorber in order to study capacity and lifetime of adsorbent.
2. The same experiment set should be conducted to remove mercury in natural gas condensate in order to compare efficiency of mercury removal.

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APPENDICES

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Appendix A

Table 1A: Condition and result of each experiment

Exp. No.	Type of mercury	Adsorbent	Temp. (C)	Hg in Feed (ppb)	Remaining Hg (ppb)
1	Mercuric chloride	-	30	984	-
2	Mercuric chloride	-	50	979	-
3	Mercuric chloride	-	75	989	-
4	Diphenylmercury	-	30	990	-
5	Diphenylmercury	-	50	985	-
6	Diphenylmercury	-	75	985	-
7	Mercuric chloride	Alumina	30	1010	170
8	Mercuric chloride	Alumina	30	987	173
9	Mercuric chloride	Alumina	30	1021	179
10	Mercuric chloride	Alumina	30	974	182
11	Mercuric chloride	Alumina	50	1010	294
12	Mercuric chloride	Alumina	50	987	297
13	Mercuric chloride	Alumina	50	1021	299
14	Mercuric chloride	Alumina	50	974	302
15	Mercuric chloride	Alumina	75	1010	459
16	Mercuric chloride	Alumina	75	987	466
17	Mercuric chloride	Alumina	75	1021	466
18	Mercuric chloride	Alumina	75	974	473
19	Diphenylmercury	Alumina	30	981	900
20	Diphenylmercury	Alumina	30	970	902
21	Diphenylmercury	Alumina	30	1034	908
22	Diphenylmercury	Alumina	30	1007	910
23	Diphenylmercury	Alumina	50	981	832
24	Diphenylmercury	Alumina	50	970	834
25	Diphenylmercury	Alumina	50	1034	838
26	Diphenylmercury	Alumina	50	1007	848
27	Diphenylmercury	Alumina	75	981	705
28	Diphenylmercury	Alumina	75	970	714
29	Diphenylmercury	Alumina	75	1034	715
30	Diphenylmercury	Alumina	75	1007	730
31	Mercuric chloride	Silica	30	1010	830
32	Mercuric chloride	Silica	30	987	830
33	Mercuric chloride	Silica	30	1021	833
34	Mercuric chloride	Silica	30	974	835
35	Mercuric chloride	Silica	50	1010	815
36	Mercuric chloride	Silica	50	987	819
37	Mercuric chloride	Silica	50	1021	821
38	Mercuric chloride	Silica	50	974	821
39	Mercuric chloride	Silica	75	1010	795
40	Mercuric chloride	Silica	75	987	798
41	Mercuric chloride	Silica	75	1021	800
42	Mercuric chloride	Silica	75	974	803
43	Diphenylmercury	Silica	30	981	960
44	Diphenylmercury	Silica	30	970	964
45	Diphenylmercury	Silica	30	1034	967
46	Diphenylmercury	Silica	30	1007	985
47	Diphenylmercury	Silica	50	981	955

Table 1A: Condition and result of each experiment (continue)

Exp. No.	Type of mercury	Adsorbent	Temp. (C)	Hg in Feed (ppb)	Remaining Hg (ppb)
48	Diphenylmercury	Silica	50	970	959
49	Diphenylmercury	Silica	50	1034	961
50	Diphenylmercury	Silica	50	1007	965
51	Diphenylmercury	Silica	75	981	937
52	Diphenylmercury	Silica	75	970	939
53	Diphenylmercury	Silica	75	1034	941
54	Diphenylmercury	Silica	75	1007	947
55	Mercuric chloride	Activated carbon	30	994	234
56	Mercuric chloride	Activated carbon	30	982	235
57	Mercuric chloride	Activated carbon	30	1007	243
58	Mercuric chloride	Activated carbon	30	989	248
59	Mercuric chloride	Activated carbon	50	994	240
60	Mercuric chloride	Activated carbon	50	982	245
61	Mercuric chloride	Activated carbon	50	1007	246
62	Mercuric chloride	Activated carbon	50	989	249
63	Mercuric chloride	Activated carbon	75	994	235
64	Mercuric chloride	Activated carbon	75	982	240
65	Mercuric chloride	Activated carbon	75	1007	240
66	Mercuric chloride	Activated carbon	75	989	245
67	Diphenylmercury	Activated carbon	30	1023	842
68	Diphenylmercury	Activated carbon	30	997	845
69	Diphenylmercury	Activated carbon	30	1017	853
70	Diphenylmercury	Activated carbon	30	985	860
71	Diphenylmercury	Activated carbon	50	1023	820
72	Diphenylmercury	Activated carbon	50	997	825
73	Diphenylmercury	Activated carbon	50	1017	834
74	Diphenylmercury	Activated carbon	50	985	837
75	Diphenylmercury	Activated carbon	75	1023	741
76	Diphenylmercury	Activated carbon	75	997	750
77	Diphenylmercury	Activated carbon	75	1017	756
78	Diphenylmercury	Activated carbon	75	985	757
79	Mercuric chloride	Silica-alumina	30	994	827
80	Mercuric chloride	Silica-alumina	30	982	831
81	Mercuric chloride	Silica-alumina	30	1007	837
82	Mercuric chloride	Silica-alumina	30	989	845
83	Mercuric chloride	Silica-alumina	50	994	737
84	Mercuric chloride	Silica-alumina	50	982	742
85	Mercuric chloride	Silica-alumina	50	1007	756
86	Mercuric chloride	Silica-alumina	50	989	765
87	Mercuric chloride	Silica-alumina	75	994	596
88	Mercuric chloride	Silica-alumina	75	982	604
89	Mercuric chloride	Silica-alumina	75	1007	610
90	Mercuric chloride	Silica-alumina	75	989	614
91	Diphenylmercury	Silica-alumina	30	1023	925
92	Diphenylmercury	Silica-alumina	30	997	930
93	Diphenylmercury	Silica-alumina	30	1017	945
94	Diphenylmercury	Silica-alumina	30	985	956
95	Diphenylmercury	Silica-alumina	50	1023	920
96	Diphenylmercury	Silica-alumina	50	997	925
97	Diphenylmercury	Silica-alumina	50	1017	931

Table 1A: Condition and result of each experiment (continue)

Exp. No.	Type of mercury	Adsorbent	Temp. (C)	Hg in Feed (ppb)	Remaining Hg (ppb)
98	Diphenylmercury	Silica-alumina	50	985	935
99	Diphenylmercury	Silica-alumina	75	1023	897
100	Diphenylmercury	Silica-alumina	75	997	901
101	Diphenylmercury	Silica-alumina	75	1017	910
102	Diphenylmercury	Silica-alumina	75	985	916
103	Mercuric chloride	Titanium oxide	30	981	689
104	Mercuric chloride	Titanium oxide	30	970	694
105	Mercuric chloride	Titanium oxide	30	1034	706
106	Mercuric chloride	Titanium oxide	30	1041	719
107	Mercuric chloride	Titanium oxide	50	981	560
108	Mercuric chloride	Titanium oxide	50	970	564
109	Mercuric chloride	Titanium oxide	50	1034	576
110	Mercuric chloride	Titanium oxide	50	1041	580
111	Mercuric chloride	Titanium oxide	75	981	389
112	Mercuric chloride	Titanium oxide	75	970	402
113	Mercuric chloride	Titanium oxide	75	1034	404
114	Mercuric chloride	Titanium oxide	75	1041	413
115	Diphenylmercury	Titanium oxide	30	963	963
116	Diphenylmercury	Titanium oxide	30	1028	970
117	Diphenylmercury	Titanium oxide	30	989	976
118	Diphenylmercury	Titanium oxide	30	1018	983
119	Diphenylmercury	Titanium oxide	50	963	958
120	Diphenylmercury	Titanium oxide	50	1028	960
121	Diphenylmercury	Titanium oxide	50	989	963
122	Diphenylmercury	Titanium oxide	50	1018	979
123	Diphenylmercury	Titanium oxide	75	963	954
124	Diphenylmercury	Titanium oxide	75	1028	959
125	Diphenylmercury	Titanium oxide	75	989	967
126	Diphenylmercury	Titanium oxide	75	1018	972
127	Mercuric chloride	CuO/alumina	30	981	96
128	Mercuric chloride	CuO/alumina	30	970	96
129	Mercuric chloride	CuO/alumina	30	1034	98
130	Mercuric chloride	CuO/alumina	30	1041	99
131	Mercuric chloride	CuO/alumina	30	997	102
132	Mercuric chloride	CuO/alumina	50	981	157
133	Mercuric chloride	CuO/alumina	50	970	162
134	Mercuric chloride	CuO/alumina	50	1034	164
135	Mercuric chloride	CuO/alumina	50	1041	165
136	Mercuric chloride	CuO/alumina	50	997	168
137	Mercuric chloride	CuO/alumina	75	981	187
138	Mercuric chloride	CuO/alumina	75	970	187
139	Mercuric chloride	CuO/alumina	75	1034	187
140	Mercuric chloride	CuO/alumina	75	1041	196
141	Mercuric chloride	CuO/alumina	75	997	198
142	Diphenylmercury	CuO/alumina	30	963	760
143	Diphenylmercury	CuO/alumina	30	1028	764
144	Diphenylmercury	CuO/alumina	30	989	775
145	Diphenylmercury	CuO/alumina	30	1018	776
146	Diphenylmercury	CuO/alumina	30	1020	780
147	Diphenylmercury	CuO/alumina	50	963	690

Table 1A: Condition and result of each experiment (continue)

Exp. No.	Type of mercury	Adsorbent	Temp. (C)	Hg in Feed (ppb)	Remaining Hg (ppb)
148	Diphenylmercury	CuO/alumina	50	1028	695
149	Diphenylmercury	CuO/alumina	50	989	704
150	Diphenylmercury	CuO/alumina	50	1018	705
151	Diphenylmercury	CuO/alumina	50	1020	716
152	Diphenylmercury	CuO/alumina	75	963	588
153	Diphenylmercury	CuO/alumina	75	1028	595
154	Diphenylmercury	CuO/alumina	75	989	602
155	Diphenylmercury	CuO/alumina	75	1018	606
156	Diphenylmercury	CuO/alumina	75	1020	612
157	Mercuric chloride	CuO/Silica	30	1010	748
158	Mercuric chloride	CuO/Silica	30	987	751
159	Mercuric chloride	CuO/Silica	30	1005	758
160	Mercuric chloride	CuO/Silica	30	990	763
161	Mercuric chloride	CuO/Silica	50	1010	689
162	Mercuric chloride	CuO/Silica	50	987	696
163	Mercuric chloride	CuO/Silica	50	1005	697
164	Mercuric chloride	CuO/Silica	50	990	706
165	Mercuric chloride	CuO/Silica	75	1010	630
166	Mercuric chloride	CuO/Silica	75	987	634
167	Mercuric chloride	CuO/Silica	75	1005	637
168	Mercuric chloride	CuO/Silica	75	990	643
169	Diphenylmercury	CuO/Silica	30	981	910
170	Diphenylmercury	CuO/Silica	30	1007	912
171	Diphenylmercury	CuO/Silica	30	1012	925
172	Diphenylmercury	CuO/Silica	30	1005	933
173	Diphenylmercury	CuO/Silica	50	981	899
174	Diphenylmercury	CuO/Silica	50	1007	902
175	Diphenylmercury	CuO/Silica	50	1012	907
176	Diphenylmercury	CuO/Silica	50	1005	912
177	Diphenylmercury	CuO/Silica	75	981	870
178	Diphenylmercury	CuO/Silica	75	1007	875
179	Diphenylmercury	CuO/Silica	75	1012	875
180	Diphenylmercury	CuO/Silica	75	1005	880
181	Mercuric chloride	CuO/Activated carbon	30	1010	110
182	Mercuric chloride	CuO/Activated carbon	30	987	110
183	Mercuric chloride	CuO/Activated carbon	30	1005	112
184	Mercuric chloride	CuO/Activated carbon	30	990	116
185	Mercuric chloride	CuO/Activated carbon	50	1010	123
186	Mercuric chloride	CuO/Activated carbon	50	987	125
187	Mercuric chloride	CuO/Activated carbon	50	1005	126
188	Mercuric chloride	CuO/Activated carbon	50	990	130
189	Mercuric chloride	CuO/Activated carbon	75	1010	134
190	Mercuric chloride	CuO/Activated carbon	75	987	137
191	Mercuric chloride	CuO/Activated carbon	75	1005	137
192	Mercuric chloride	CuO/Activated carbon	75	990	140
193	Diphenylmercury	CuO/Activated carbon	30	981	700
194	Diphenylmercury	CuO/Activated carbon	30	1007	704
195	Diphenylmercury	CuO/Activated carbon	30	1012	708
196	Diphenylmercury	CuO/Activated carbon	30	1005	712
197	Diphenylmercury	CuO/Activated carbon	50	981	710

Table 1A: Condition and result of each experiment (continue)

Exp. No.	Type of mercury	Adsorbent	Temp. (C)	Hg in Feed (ppb)	Remaining Hg (ppb)
198	Diphenylmercury	CuO/Activated carbon	50	1007	715
199	Diphenylmercury	CuO/Activated carbon	50	1012	721
200	Diphenylmercury	CuO/Activated carbon	50	1005	722
201	Diphenylmercury	CuO/Activated carbon	75	981	684
202	Diphenylmercury	CuO/Activated carbon	75	1007	684
203	Diphenylmercury	CuO/Activated carbon	75	1012	689
204	Diphenylmercury	CuO/Activated carbon	75	1005	695
205	Mercuric chloride	CuO/Silica-alumina	30	991	710
206	Mercuric chloride	CuO/Silica-alumina	30	997	713
207	Mercuric chloride	CuO/Silica-alumina	30	1008	714
208	Mercuric chloride	CuO/Silica-alumina	30	1011	719
209	Mercuric chloride	CuO/Silica-alumina	50	991	530
210	Mercuric chloride	CuO/Silica-alumina	50	997	530
211	Mercuric chloride	CuO/Silica-alumina	50	1008	532
212	Mercuric chloride	CuO/Silica-alumina	50	1011	536
213	Mercuric chloride	CuO/Silica-alumina	75	991	410
214	Mercuric chloride	CuO/Silica-alumina	75	997	416
215	Mercuric chloride	CuO/Silica-alumina	75	1008	418
216	Mercuric chloride	CuO/Silica-alumina	75	1011	424
217	Diphenylmercury	CuO/Silica-alumina	30	1019	861
218	Diphenylmercury	CuO/Silica-alumina	30	1025	867
219	Diphenylmercury	CuO/Silica-alumina	30	1009	869
220	Diphenylmercury	CuO/Silica-alumina	30	997	871
221	Diphenylmercury	CuO/Silica-alumina	50	1019	819
222	Diphenylmercury	CuO/Silica-alumina	50	1025	820
223	Diphenylmercury	CuO/Silica-alumina	50	1009	820
224	Diphenylmercury	CuO/Silica-alumina	50	997	833
225	Diphenylmercury	CuO/Silica-alumina	75	1019	790
226	Diphenylmercury	CuO/Silica-alumina	75	1025	794
227	Diphenylmercury	CuO/Silica-alumina	75	1009	799
228	Diphenylmercury	CuO/Silica-alumina	75	997	801
229	Mercuric chloride	CuO/Titanium oxide	30	991	538
230	Mercuric chloride	CuO/Titanium oxide	30	997	542
231	Mercuric chloride	CuO/Titanium oxide	30	1008	544
232	Mercuric chloride	CuO/Titanium oxide	30	1011	552
233	Mercuric chloride	CuO/Titanium oxide	50	991	457
234	Mercuric chloride	CuO/Titanium oxide	50	997	466
235	Mercuric chloride	CuO/Titanium oxide	50	1008	469
236	Mercuric chloride	CuO/Titanium oxide	50	1011	472
237	Mercuric chloride	CuO/Titanium oxide	75	991	245
238	Mercuric chloride	CuO/Titanium oxide	75	997	251
239	Mercuric chloride	CuO/Titanium oxide	75	1008	259
240	Mercuric chloride	CuO/Titanium oxide	75	1011	261
241	Diphenylmercury	CuO/Titanium oxide	30	1019	930
242	Diphenylmercury	CuO/Titanium oxide	30	1025	932
243	Diphenylmercury	CuO/Titanium oxide	30	1009	940
244	Diphenylmercury	CuO/Titanium oxide	30	997	954
245	Diphenylmercury	CuO/Titanium oxide	50	1019	907
246	Diphenylmercury	CuO/Titanium oxide	50	1025	912
247	Diphenylmercury	CuO/Titanium oxide	50	1009	916

Table 1A: Condition and result of each experiment (continue)

Exp. No.	Type of mercury	Adsorbent	Temp. (C)	Hg in Feed (ppb)	Remaining Hg (ppb)
248	Diphenylmercury	CuO/Titanium oxide	50	997	925
249	Diphenylmercury	CuO/Titanium oxide	75	1019	869
250	Diphenylmercury	CuO/Titanium oxide	75	1025	874
251	Diphenylmercury	CuO/Titanium oxide	75	1009	887
252	Diphenylmercury	CuO/Titanium oxide	75	997	890



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Appendix B**Table 1B:** Properties of toluene*

Formula	C ₇ H ₈
Chemical Name	Toluene
Physical Properties	
Molecular Weight	92.13
Form	Liquid
Color	Colorless
Boiling Point (°C)	110.8
Melting Point (°C)	-95
Specific Gravity	0.866
Solubility	soluble in ether and alcohol

*From MSDS

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Table 2B: Properties of Mercuric chloride*

Formula	HgCl ₂
Chemical Name	Mercuric chloride
Physical Properties	
Molecular Weight	271.52
Form	solid
Color	white
Boiling Point (°C)	302
Melting Point (°C)	277
Specific Gravity	5.44
Solubility	soluble in water

*From MSDS



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Table 3B: Properties of Diphenylmercury*

Formula	C ₁₂ H ₁₀ Hg
Chemical Name	Diphenylmercury
Physical Properties	
Molecular Weight	354.8
Form	solid
Color	white
Boiling Point (°C)	-
Melting Point (°C)	121-124
Specific Gravity	2.32
Solubility	moderately soluble in toluene
Purity	>97%
Supplier	Fluka

*From Supplier

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Table 4B: Properties of Aluminum Oxide, activated, neutral Brockmann*

Formula	Al_2O_3
Chemical Name	Neutral Alumina
Physical Properties	
Form	solid
Color	white
Standard grade	150 mesh
Surface area	155 m^2/g
pH of aqueous suspension	7.0 \pm 0.5
Supplier	Aldrich

*From Supplier



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Table 5B: Properties of Copper Nitrate*

Formula	$\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$
Chemical Name	Copper nitrate trihydrate
Physical Properties	
Molecular Weight	241.6
Form	solid
Color	blue
Boiling Point (°C)	-
Melting Point (°C)	114.5
Specific Gravity	2.32
Solubility	soluble in ether and alcohol
Purity	>99%

*From Merck Index

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Table 6B: Properties of Nitric Acid*

Formula	$\text{H}_2(\text{NO}_3)_2$
Chemical Name	Nitric acid
Physical Properties	
Molecular Weight	63.02
Form	liquid
Color	colorless
Boiling Point (°C)	86
Melting Point (°C)	-41.59
Specific Gravity	1.502
Solubility	soluble in water
Purity	69.0-70.597%
Supplier	Merck

*From Merck Index

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Table 7B: Properties of Hydrochloric Acid*

Formula	HCl
Chemical Name	Hydrochloric acid
Physical Properties	
Molecular Weight	36.47
Form	liquid
Color	colorless
Melting Point (°C)	-15.35
Specific Gravity	1.05
Solubility	soluble in water
Purity	37%
Supplier	Merck

*From Supplier

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Table 8B: Properties of Sulfuric Acid*

Formula	H ₂ SO ₄
Chemical Name	Sulfuric acid
Physical Properties	
Molecular Weight	97.09
Form	liquid
Color	colorless
Specific Gravity	2.03
Solubility	soluble in water
Purity	95.7%
Supplier	Merck

*From Supplier



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Table 9B: Properties of Hydrogen Peroxide*

Formula	H ₂ O ₂
Chemical Name	Hydrogen peroxide
Physical Properties	
Molecular Weight	34.02
Form	liquid
Color	colorless
Specific Gravity	1.13
Solubility	soluble in water, acid and ether
Purity	33%
Supplier	Carlo Erba

*From Supplier

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Table 10B: Properties of Potassium Permanganated*

Formula	KMnO ₄
Chemical Name	Potassium Permanganate
Physical Properties	
Molecular Weight	158.03
Form	solid
Color	dark purple
Specific Gravity	2.71
Solubility	soluble in water
Purity	>99%

*From Merck Index



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Table 11B: Properties of Potassium Persulfate*

Formula	K_2SO_8
Chemical Name	Potassium Persulfate
Physical Properties	
Molecular Weight	270.32
Form	solid
Color	white
Solubility	soluble in water
Purity	>99%

*From Merck Index



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Table 12B: Properties of Hydroxylamine Hydrochloride*

Formula	$\text{NH}_2\text{OH}\cdot\text{HCl}$
Chemical Name	Hydroxylamine Hydrochloride
Physical Properties	
Molecular Weight	69.49
Form	solid
Color	white
Boiling Point (°C)	58
Melting Point (°C)	33
Solubility	soluble in water
Purity	>99%

*From Merck Index



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