

A LITERATURE REVIEW ON THE POSSIBLE SOURCES FOR PRODUCTION OF SULPHURIC ACID

Nabihah binti Mohammad, Dr Sharif Abdulbari Ali

Faculty of Chemical Engineering, Universiti Teknologi Mara, Shah Alam

Abstract—Sulphuric acid is a universal chemical or substances that mostly used in various applications in the world. There are many uses of sulphuric acid which including catalyst, solvent, cleaning agent and others. The purpose of this research is to review three possible sources of production of sulphur dioxide for sulphuric acid manufacturing and determine most feasible source to be manufactured commercially. The impacts of sulphuric acid to the human were determined by a review of the past case study. It was found that elemental sulphur is the most feasible source of sulphur dioxide for sulphuric acid manufacturing commercially. The impacts of sulphuric acid to the human also being proved by the past incident occur around the world.

Keywords— Sulphuric Acid, Sulphur Dioxide, Possible Sources, Feasible

I. INTRODUCTION

Sulphuric acid is an important industrial chemical as it is one of the most used and universal material/ chemical substance in various industries. It is widely used in the manufacturing processes of many goods over an extensive range of applications. The major applications of sulphuric acids includes in the production of fertilizers and manufacture of chemicals [1]. There are also some growing number of ends-uses and applications using this sulphuric acid. So obviously the demand on the sulphuric acid is increasing year by year. As we know, there is various type of manufacturing process for sulphuric acid. But, there is still having some limitations of the process on-going. So, the processes will always being improved in order to get better result as the world demand was increased.

There will be some disadvantages of the process that have been carried out in early years of discovery. So the improvement is carrying out in stages. For example, sulphuric acid was produced at Nordhausen from green vitriol but it was expensive. So, the improved process for its synthesis by burning sulphur with saltpeter (potassium nitrate). It was first used by Johann Glauber in the 17th century and developed commercially by Joshua Ward in England c.1740. After that, it was soon superseded by the lead chamber process, invented by John Roebuck in 1746 and since improved by many others. Then, among the abundant existence of manufacturing process, there will be their specific improvement that has been done for each and every upgraded process. So, there will some needs to find and analyse the relevant process for industrial used that will give more advantages among others [2].

The common problems or issues arise for sulphuric acid manufacturing includes the increment of capital and operating cost whenever the production rate was increased. As we know, this is for sure will happen because, to increase the production rate there will be some needs to increase on the capacity of raw materials and the number/size of machinery involves. But the problem can be solved if some improvements were made. For instance, the usage of catalyst into the process or changing operating conditions to the more favourable and optimum one. Other than that, the problem exist is maximizing production rate will increase the wastes in the forms of solids, liquids or gas. The waste produced will absolutely create more cost for the waste treatment before discharge. Another point is the upgraded technology/process is not feasible to commercialize in industrial scale, it is just suitable for laboratory scale only. The last one is about environmental concerns. The potential gas created in this manufacturing process can cause serious air pollutants at quite low concentrations. The gas involves are sulphur dioxide and hydrogen sulphide that caused by oxidation and reduction process of the sulphur itself.

Consequently, these conditions absolutely increase in environmental concern as a result of the on-going environmental deterioration for future generations. Unfortunately, various efforts to eliminate environmentally destructive behaviour through some interventions have typically not successfully conducted. However, it showed a positive effect in reducing the behaviors that damage the environment government agencies, companies and the public.

This research was conducted to to determine three possible sources of production of sulphur dioxide for sulphuric acid manufacturing and determine most feasible source to be manufactured commercially. The impacts of sulphuric acid to the human were determined by a review study.

II. METHODOLOGY

A. Materials and Method of Extracting Data

There are various of information explaining related to the sulphuric acid industry. The available data found in the numerous reading tools including the history of the production of sulphuric acid, the application of sulphuric acid industry, the demand of this universal chemical substances and many other information related. All of the useful data obtained from the trusted sources were divided to the suitable research sections. The useful reading materials available including book, journal, patent, conference, internet websites and report. These materials can be obtained by surfing the internet or looking through the library.

The first sources are from book. There are many books from different writers and publishers are available online and even offline. The related books can be reaching either by online reading (e-book) or it also can be downloaded as copy for future reference. Looking through the different books, they are actually giving the same or nearly the same information but different in how it's elaborated. But, there will be some added information for the latest edition of the books. The contents of each book will be different as the different title of the books. For example, the book with title 'Sulphuric acid industry' will present more on the application of the sulphuric acid while the book with the title 'handbook of sulphuric acid manufacturing' will be present the data on the process of manufacturing sulphuric acid. The title of the books also plays role in order to search the right data for the research. So, in order to get enough information for the research paper, the data from several books will be collected then it will be analyzed in order to make sure that only the suitable, required and logic information will be written in the research paper.

Other than that, journal and article also used in the data searching in order to complete this research review paper. There are many types of online and offline journal that are available. It was including journal by Elsevier, article by national pollutant inventory, environmental protection agency and many others. Among all those information obtained, not all of them were suitable to be written in the research paper. So, the data were analyzed first in order to ensure the conformity of the extracted data from those journal and article. The data usually extracted from the readings materials by using method of general to specific. Meaning that, the data was obtained in general form first, and then it will be specified according to the research objectives.

Besides, patent also important in getting and obtaining the precise and accurate data for writing a research paper. The data related to the sources of the sulphur dioxide for sulphuric acid manufacturing can be obtained by referring to the patent by several countries. Patents also precisely explained on the two commercially used method of sulphuric acid manufacturing in details. But, for this research paper, the details of the sulphuric acid production process are not needed, but it is only written on the general one as extra information only. This is because the research objective is on the sources of the sulphur dioxide, raw materials of sulphuric acid manufacture not about the production of sulphuric acid.

All of the related data obtained from various sources of reading materials were then specify in order to ensure the originality and the authenticity of the sources. The sources and references of the information obtained are based on the most usually referred by researcher or writer and also authentic. Besides, the chosen sources for obtaining information and data is based on the validity of the sources. For example, the first source which is from the website give the information for the first explored of the sulphuric acid is on 8th century by Jabir Ibn Hayyan. Then, the crosschecked of the information is carried out at another authentic site. If the given information at the authentic site or publication is nearly the same with the previous website, so the source is valid to refer. Otherwise, the source will not valid to be referred.

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PROGRESS REPORT FORM FOR FINAL YEAR RESEARCH PROJECT 1 (CHE687 / CHE694)
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Name: Nabihah Binti Mohammad
ID: 201305099
Supervisor: Dr. Sheriff Abdullah Ali
Title: A Literature Review On The Possible Sources of Production Of Sulphuric Acid

Summary of Journal

TITLE	YEARS / AUTHOR	SOURCE / NAME OF JOURNAL	PROBLEM STATEMENT	TECHNIQUE / METHODOLOGY	REMARK/FINDINGS	CONCLUSION & RECOMMENDATION
Sulphuric acid production via Chemical Looping Combustion of elemental sulphur	Francisco Garcia-Laborda, Luis F. de Diego, Arturo Caballero, Pilar Gayán, Alberto Abad, Juan Adalberto, Gerald Strachmann b (2016)	ELSEVIER, Applied Energy	-The presence of sulphur in fuel is normally detrimental for many oxygen carriers based (NiO or Cu ₂ O) -production of nitrogen oxides gas depends on temperature in the air reactor	-burning elemental sulphur in a 500Wth CLC unit with a high sulphur-resistant oxygen carrier based on iron oxide -elemental sulphur combustion by an oxygen carrier	Complete combustion of sulphur to SO ₂ was achieved and no destruction of the oxygen carrier was observed.	the production of H ₂ SO ₄ via CLC is a feasible technology which can provide a series of relevant techno-economic benefits in comparison with current industrial production process
Third Journal						
Fourth Journal						

Figure 1: Tabulated data from various reading sources

B. Step in Conducting A Systematic Review

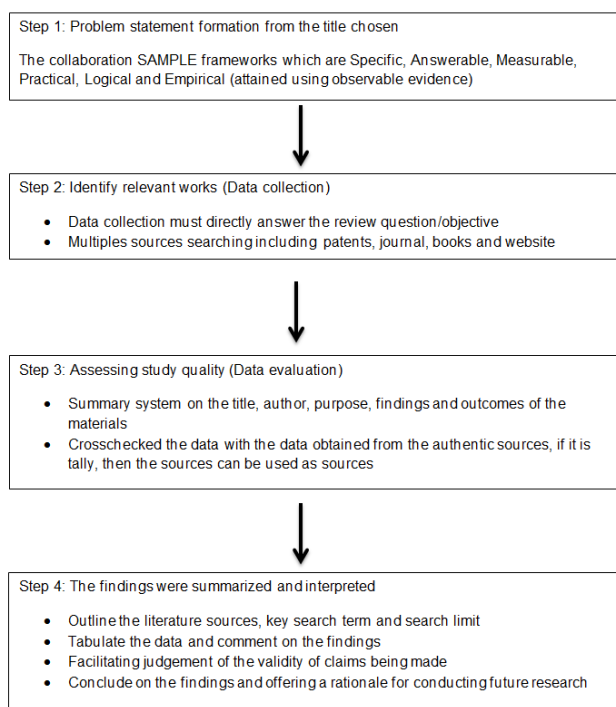


Figure 2: Step in conducting a systematic review paper [3]

III. RESULTS AND DISCUSSION

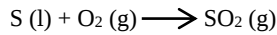
A. Possible sources of production of sulphur dioxide for sulphuric acid manufacturing

There are three possible sources of sulphur dioxide for production of sulphuric acid for industrial use. The percentage of total supply was presented as in table 1.

Table 1: Sources of sulphur and SO₂ for sulphuric acid production [4]

Sources of SO ₂	Percentage of total supply (%)
Elemental sulphur	70
Smelting & Roasting gases	20
Decomposing spent petroleum	10

I. Production of sulphur dioxide from elemental sulphur



Elemental sulphur can be oxidized by hydrogen peroxide to form sulphur dioxide. Then it will further oxidized to form sulphur trioxide. After all, it will be reacts with water to form sulphuric acid. This process thermodynamically feasible but it quite slow. The elemental sulphur will first melt, and then it will spray into an excess of dry air at atmospheric pressure. Other than that, burning elemental sulphur also constitute one of the way to produce sulphur dioxide. All of this sulphur is virtually the by-product of natural gas and petroleum refining. The product details of this burning elemental sulphur are written in the table 2.

Table 2: Product details of burning elemental sulphur [5]

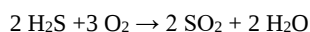
Products	Percentage by volume (%)
Sulphur dioxide (SO ₂)	~12
Oxygen (O ₂)	9
Nitrogen (N ₂)	79

This composition is perfect for further process which catalytic sulphur dioxide oxidation then sulphuric acid processing. So, it eases the step for sulphuric acid manufacturing. After burning, it can be proceed to further process as soon as possible without any intermediate process.

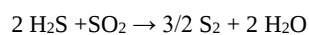
Elemental sulphur can be obtained by various methods including the sulphur recovery technologies, by Claus process and also extracted sulphur by using Frasch process. Claus process is the most common conversion method used in order to recover the elemental sulphur. This process was first patented in 1883 by the chemist Carl Friedrich Claus, the Claus process has become the industry standard. It is because, sulphur recovered and produced by using this process was approximately 90% to 95% of total [6]. The Claus process has been known and used in the industry for over 100 years. In this process, hydrogen sulphide, H₂S which a by-product of processing natural gas will be convert to the elemental sulphur by specific condition.

Claus process is using the reaction between hydrogen sulphide and sulphur dioxide (produced in the Claus process furnace from the combustion of H₂S with air and/or oxygen) yielding elemental sulphur and water vapour. The reaction in the Claus process is consisting of two steps. The reactions involved were as follows: [7]

First step:



Second step:



According to the American journal of environmental science, this technology process can possibly provide about 96-97% conversion of influent sulphur in H₂S to S [7]. Majority of about 92% of total sulphur (8 million metric tons) produced in the United States in 2005 while in 2010, the vast majority of the 68 million metric tons of sulphur produced worldwide was recovered by industrials by-products using this process. The table 3 provide the typical analyses of the Claus feed gases.

Table 3: Feed gas composition [8]

Typical feed gas composition					
Component	Mol %	Wt %	Component	Mol %	Wt %
From an amine process:			From a sour water stripper:		
H ₂ S	82.1	80.8	H ₂ S	26.7	40.2
CO ₂	11.9	15.1	CO ₂	2.6	5.1
NH ₃	nil	nil	NH ₃	39.4	29.7
H ₂ O	4.0	2.1	H ₂ O	31.3	25.0
HC	2.0	2.0	HC	nil	nil

Notes: H₂S is hydrogen sulfide, CO₂ is carbon dioxide, NH₃ is ammonia, H₂O is water and HC is an abbreviation for hydrocarbons. Wt % is weight percent .

The gases which consisting of over 25% of composition H₂S are suitable for recovery of sulphur through Claus process while for the gases that having lesser amount of H₂S will proceed with other process design configurations [8]. The sulphur recovery by Claus process consist of three main steps which including:

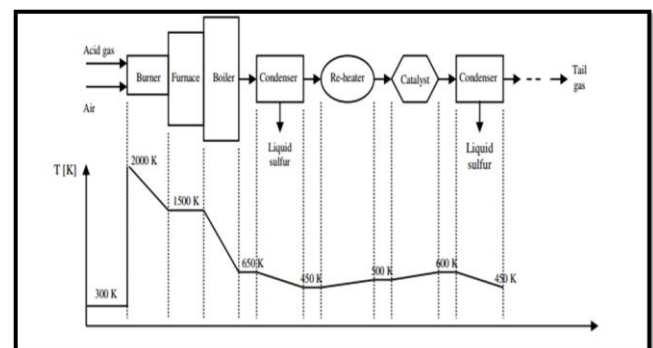


Figure 3: Process flow diagram of a Claus process [9]

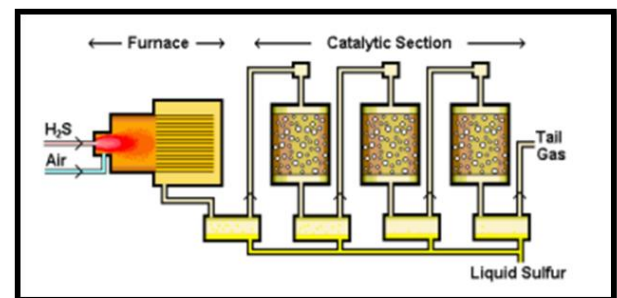


Figure 4: Arrangement of a Claus unit [10]

However, with the increasing world demand of sulphur, the Claus process goes through various improvements in order to face their limitations and satisfy the higher contents of sulphur. New technology was introduced and investigated in order to improve the sulphur recovery to a higher production. Unfortunately, from the previous study, all improvements to satisfy very high sulphur recovery created very high cost addition. However, the modified Claus process was greatly increased the sulphur yield and used as the basis of most Sulphur Recovery Units today [11].

Move to the second process which is Frasch process. Frasch process constitutes a method in which sulphur is extracted from naturally occurring elemental deposits of sulphur. This method of mining deep-lying sulphur was invented by a chemist originally from American, Herman Frasch. It is the main commercial source of sulphur. It is because, this Frasch process is the only economic method of recovering sulphur from elemental deposits. The Frasch process make of use of a steel tube that made up of three concentric pipes drilled up to the beds of sulphur which is in the range of 500 to 3000 feet deep. This sulphur produced is contains of minimum purity of 99.5% percent. So it is very suitable to use in sulphuric acid manufacturing and also for majority use in all industry. The illustration of this process was as follows in figure 5:

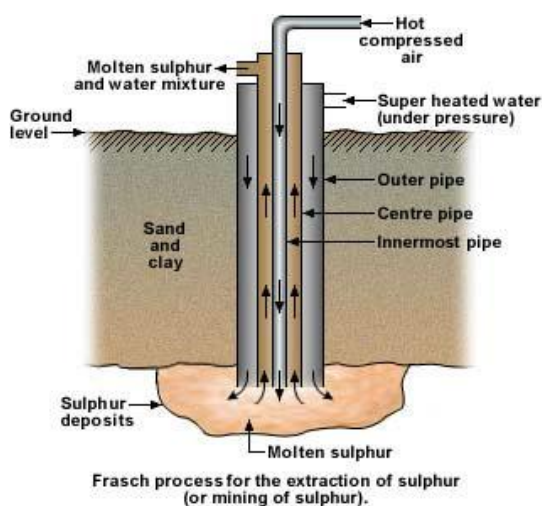


Figure 5: Frasch process for the extraction of sulphur [12]

The process was begun by pumping down the superheated water under great pressure in the pipe. The purpose of doing this step is to melt the sulphur deposited. It will flows to the middle tube. However, the water pressure is insufficient in order to push the molten sulphur into the surface of the ground. It is due to the greater density of the molten sulphur. So, the compressed air was injected via the middle pipe creating it less dense. Then, the mixture of sulphur (molten) and water will force up by the innermost pipe and discharged into bins. The mixture of water and sulphur was then separated and the sulphur is going to be solidified in order to attain the pure elemental sulphur.

Some dissolved minerals will be exists in the recovered water resulted from this extraction process. In order to make sure that these things is not affect our environment, the effluent need to going through some small treatment. The effluent should undergo cooling process in order to avoid thermal pollution. It is also need to be recycled to avoid contamination of local ecosystem. The elemental sulphur recover by using this process having a high purity will can go up to 99% which is suitable for majority uses in industry.

The melting point of sulphur is quite low which roughly about 115 °C due to it weak dispersion forces. This property making it will easily melts by superheated water. The nature of sulphur which it is insoluble in water and it also does not react chemically with water allows the separation process of water and sulphur easy and economic. Other than that, sulphur-water emulsion is a light compound as sulphur having a low density properties which is 2.07

g/cm³ resulting on the easily ready to be transported to the surface using the compressed air. This process also requires no special precautions in order to protect the health of miners. It is because, the sulphur's properties itself which are inert, non-toxic and non-volatile. So, there were no additions of costs for special precautions [13].

For the sources of sulphur dioxide that obtained from the elemental sulphur, the processing types that most frequently used and more preferable is the Claus process instead of Frasch process. This is because, there are many difficulties arise while removing the contaminants in the Frasch process. [14]

II. Production of Sulphur dioxide from smelter and roaster offgases

As stated in the table 1.1, about 20% of the world's sulphuric acid is made from sulphur dioxide in smelter and roaster offgases. Volume of sulphur dioxide content in these gases is around 10% – 75% [15]. This type of sulphur dioxide sources is very suitable for making sulphuric acid. But there are some contaminants that are attached with the required gas. So, the gases must go through some steps before acid making. The steps are as in figure 6.



Figure 6: Preparation steps before sent to acid making [16]

There are several metal smelting and roasting that producing and releasing the gases that is required in order to manufacture sulphuric acid. The gas mentions is the sulphur dioxide. As for information, sulphur dioxide constitutes the main raw material that is used to produce sulphuric acid. The recovery of sulphur dioxide from metal smelting and ore roasting is including primary copper smelting, roasting of metal ores (Galena and Sphalerite), roasting of pyrites and others.

The first one is on primary copper smelting. The past incident on this copper smelter is about the sulphur dioxide that released to the atmosphere. The US sources had released millions tons of sulphur dioxide to the atmosphere in 1970 and 10.6% of that total is emitted from the primary copper smelter which a significant number of emissions [17]. The copper smelters roast the dried concentrates before smelting. The sulphur is being oxidized and the volatile metal impurities escape while roasting. The oxidized sulphur is released as sulphur dioxide which is needed in making sulphuric acid. Roasting improves the efficiency of smelting as it

reduces the water content in the materials. The molten copper matte will be going through some reaction in order to get the copper product that will be marketed. It undergoes a series of chemical reactions for the purpose of removing remaining impurities. This stage is to obtain 97-99% of pure copper.

Besides, there are two sulphides metal ores that usually found together which are Galena and Sphalerite. Galena constitutes a lead sulphide mineral while Sphalerite is a zinc sulphide. The chemical formulas of these two metal ores are:



Figure 7: Chemical formula of Galena and Sphalerite

As for information, the world's primary ore of lead is Galena. It is very easy to identify which it exhibit perfect cleavage in three directions that intersect at 90 degrees. The colour of this substance is bright silver. The chemical structure of galena composed of equally number of lead and sulphide ions. The typical composition of galena is about 86% lead and 14% sulphur by weight. The ions arrangement is in a cubic pattern. The arrangement is shown in figure 8.

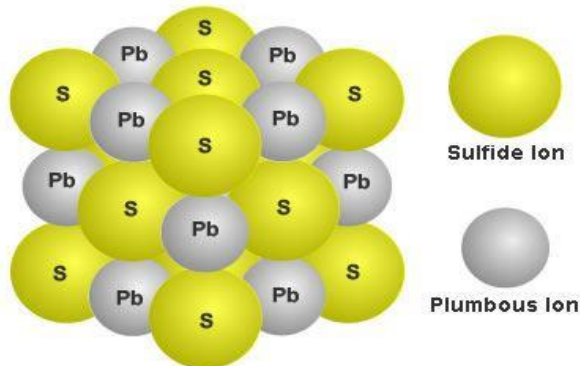


Figure 8: Structure of galena (ions arrangement)[18]



Figure 9: Photograph of a nice cubic galena crystal with adjacent calcite crystals [18]



Figure 10: Cleavage fragments of galena [18]

For the second metal ore which is sphalerite, it occurs in various colours and forms. The previous name used for this mineral including zinc blende, blackjack and others. The zinc sulphide contains many impurities including iron, magnesium and sometimes with manganese. The chemical formula of sphalerite is (Zn, Fe)S. This is because the zinc sulphide containing variable amounts of iron. But then, the amount of iron is usually less than 25% by weight [18]. It has a total of six cleavages of directions [19]. This type of mineral is difficult to identify than other mineral ores types such galena. The structure of the sphalerite is as though to the diamond structure. The sphalerite's structure is shown as in figure 11. The colour of sphalerite is differ for different origin.



Figure 11: Sphalerite with dolomite from Gilman, Colorado [18]



Figure 12: Sphalerite from Ontario, Canada [20]



Figure 13: Sphalerite from Wallis, Switzerland [20]



Figure 14: Sphalerite from Tuscany, Italy [20]

However, these two types of mineral ores were not much contributed to the creation and emission of the sulphur dioxide. The sulphur dioxide gases emitted from smelting and roasting activities is only 20% of the total sulphur dioxide obtained for the sulphuric acid making for industrial used.

III. Production of Sulphur dioxide from decomposing spent petroleum catalyst

Secondary materials which resulting from other processes is important source for metals to meet future growing requirements in chemical industries. It is due to the exhaustion of the high grade ore and also difficulty in obtaining low grade ore. The secondary resource that play significant role is the spent catalyst. It is because this type of secondary resources related to issues of disposal of huge amounts of recoverable and valuable metals. So, the spent catalyst is regarded as waste when these catalysts lose their activity or selectivity. There are probable reasons for the deactivation of the catalyst. The reasons are including [21]:

- 1) Overheating
- 2) Deposition of carbon and sulphur
- 3) Contamination with heavy metals (lead, arsenic and vanadium)

This waste created few environmental issues if it is not go through treatment before being disposed. So, the spent catalyst require a treatment process in order to make sure that it is pollution free and the valuable metals could be recover. Table 4 shows the contents of spent catalyst that were usually used in the petroleum industry.

Table 4: Percentages of the spent catalyst element's content [21].

Elements	Percentage (%)
Molybdenum	4 – 12
Aluminium	15 – 30
Nickel	1 – 5
Cobalt	0 – 4
Sulphur	5 – 10
Silicon	1 – 5
Vanadium	0 – 0.5

In order to increase the efficiency of recovery of metals from spent catalysts, variety of processing approaches has been proposed. A generous attention has been bringing into focus on hydro-metallurgical process which includes water/acid leaching come behind the salt roasting. For the purpose to yields better efficiency of metal extraction, the treatment of the sulphide based spent catalyst by using high temperature roasting process is used instead of the previous processing treatment.

However, the recovery sulphur that will be obtained in this track is not much. It is due to the originally low sulphur content in the spent catalyst. It only can recover in the range of 5 – 10 % of sulphur dioxide. So, this source of sulphur dioxide is not recommended to use for sent to acid making process to manufacture sulphuric acid. This process is more suitable to recover other metals in order to meet the future growing requirements in steel and chemical industries.

B. The impacts of sulphuric acid to the human

Sulphuric acid and sulphur dioxide constitute a harmful and hazardous substance. According to the agency for toxic substances and disease registry, it is one in series of Public Health Statements about hazardous and their health effects. However, the effects of the exposure to any hazardous substances or elements are depending on few factors. The factors and aspects that could be included into consideration are the dosage the duration, how you are exposed, personal traits and habits, and whether other chemicals are present [22]. However, if the concentrated sulphuric acid comes into contact, it is highly corrosive to all tissues.

The major routes of exposure to the sulphuric acid including inhalation, ingestion, dermal and ocular exposure. The exposure to the sulphuric acid is divided into two types whether acute exposure or chronic exposure. Acute exposure means immediately after the exposure. The meaning of an acute exposure is a result to a high level exposure over a short period of time. While, chronic exposure is occurring over a longer period. Table 5 shows the signs or symptoms of the exposure to the sulphuric acid by each respectively routes.

Table 5: The signs or symptoms of the exposure to the sulphuric acid by each respectively routes[23].

Route	Signs and symptoms
Inhalation	• Irritation of the eyes and nose • Sore throat • Cough and difficulty breathing • Chest tightness • Headache and fever • Corrosive damage to the mucous membranes
Ingestion	• Immediate pain • Burning in mouth, throat and stomach • Vomiting, haematemesis and dyspnoea • Oedema may cause swallowing difficult • Acids can cause damage to the stomach with ulceration
Dermal	• May cause blistering, ulceration and penetrating necrosis • Coagulation burns may develop with the destruction of the surface epithelium and sub-mucosa

Human may be exposed to the sulphuric acid at various places which including workplace, home, or environment itself (near to hazardous waste site). Sulphuric acid is a universal chemical that being used in various applications. So, every workplace that involved in the usage of it will be having risk. However, the exposure level is different depend on the type of workplace. The one that work with the chemical and metal plating industry, production of detergents, soaps or fertilizers industry may be exposed to the sulphuric acid.

Other than that, people may also be exposed to this substance by breathing outdoor air. The greater risk of exposure by breathing is for the people who live near to the hazardous waste site. Accidental spills of sulphuric acid also one of the way of exposure. But, high probability of these accidents to occur is at site. So, for reducing the risk of exposure, EPA had limits the amount of sulphur dioxide that can be released to the air. OSHA limits the amount of sulphuric acid that can be present in workroom air to 1 mg/m³ while NIOSH also recommends a time-weighted average limit of 1 mg/m³ [22].

The past incident cases proves that the sulphuric acid give different effect to human health as the different routes of exposure. The first industrial case is by inhalation of sulphuric acid fume by a 40-year old worker. The worker was accidentally sprayed the sulphuric acid fumes in the face resulting an acute exposure effect to him. He had acute respiratory symptoms which permanent pulmonary damage. It was categorized by chronic cough and difficulty in breathing. Other than that, he also suffers in bronchiectasis with fibrosis, emphysema developing and also reducing in performance of respiratory. The period of suffers of all this effects is within a 7-18 month period [22]. In another industrial case, the test was carried out for workers that were exposed to the sulphuric acid for an average of 12.2 years. A small decrease in forced vital capacity (FVC) was detected in workers which exposed to an average sulphuric acid aerosol concentration of 0.21 mg/m³ compared to workers exposed to an average concentration of 0.1 mg/m³. There was no other serious changes in lung function tests were noted.

The first case of the past incident of ingestion of sulphuric acid was occurred in 1825. An adult having swallowed by mistakes of two ounces of concentrated sulphuric acid, she was survive but remained a long time affected by consequences of cauterization by the acid. The second case is in 1835, an adult aged 27 wished to

poison herself by swallowing an amount of concentrated sulphuric acid [24]. Fortunately, someone found her wrong doing and immediately save her. She has been treated and survived. In another case, a man splashed over the face and body with a solution containing sulphuric acid developed second-degree burns over 60% of his body and third-degree burns over 20% of his body [25]. He died 5 days later from the extensive burns and chemical damage to the respiratory tract. The last but not least was incident that occurred in North Carolina which an employee of a janitorial supply store was filling an overhead storage tank with a bucket of sulphuric acid when the ladder slipped, causing him to spill five gallons of acid on his body. He was admitted to the hospital with second-degree chemical burns over 40 percent of his body [26].

IV. CONCLUSION

Sulphuric acid industry has been growing day by day resulting in increased production capacity in the world rankings. So, it is significant to find the possible sources of the production of sulphur dioxide which is the raw materials in manufacturing sulphuric acid chemical. As the research has demonstrated, the three possible sources of production of sulphur dioxide for sulphuric acid manufacturing has been discovered are elemental sulphur, smelting and roasting ores and from decomposing spent catalyst in petroleum industry. While, the most feasible and suitable source for the sulphuric acid manufacturing in commercial is by using elemental sulphur as it produced the largest amount among others and also meet the criterions required. The last but not least, the objective on the impacts of the sulphuric acid to the human was successfully determined and proved by the past incident occurs in the world.

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