1. Introduction

Sulphur is one of the world’s common chemical elements that occur naturally in its native form, a solid crystal substance, or extracted mainly from petroleum refineries and natural gas processing, thus creating a global surplus of sulphur [1]. Sulphur as a non-metals is capable of combining essentially with all chemical elements and form various compounds; such as sulphuric acid, produced as a by-product of ferrous and non-ferrous metal smelting. Other compounds, such as sulphur dioxide, come from the emission from petroleum products used in cars and coal generating electricity. Its other properties include low thermo- and electro-conductivity and low water solubility.

**Elemental sulphur (Brimstone):** This refers to sulphur produced in its elemental form, that is, sulphur mined by processes such as Frasch or as native refined sulphur, or recovered from gas and oil production [2]. Elemental sulfur is mostly used in the production of sulfuric acid for the preparation of fertilizers. In addition, it has been also employed as a concrete additive and more recently, lithium-sulfur batteries and polymeric materials [3].

**Lime sulphur:** Lime sulphur, a mixture of calcium and sulphur (calcium polysulfide), has been used as a pesticide to control diseases in a wide range of plant species for over a century in both floriculture and horticulture. Firstly, lime sulphur dips are used in veterinary industry to cure dogs, cats, puppies and kittens suffering from non-specific dermatoses and parasites [4].

**Sulphur in other forms (SOF):** The forms other than brimstone or pyrites, where sulphur is produced in the form of sulphuric acid or other sulphur compounds without prior refining to brimstone. SOF, mainly, originates from the smelting of non-ferrous metals, led by copper, followed by zinc, lead, and nickel [5].

**Sulphur in all forms:** The aggregate of brimstone, the equivalent sulphur content of pyrites, and the equivalent SOF [5].

**Discretionary (voluntary) and non-discretionary (non-voluntary) sulphur:** where there is an economic case for producing, Sulphur can also be classified as discretionary sulphur which normally includes Frasch, native sulphur, and pyrites. Non-discretionary sulphur has to be recovered, regardless of the economics of sulphur recovery, either for environmental/regulatory or process reasons. Recovered sulphur and SOF are almost always non-discretionary [5].
The oil and gas industry must look at sulphur as a primary product and manage its production, and a large quantity of sulfur is obtained from the current environmental restrictions regarding the petroleum and gas refining processes, coal processing, and refining of copper in the mining sector. Thus, the interest appeared regarding the application of sulphur in different fields, and extensive research programs initiated with the focus on various properties of the material. Sulphur is utilized in many fields the most important of which are agriculture, construction and energy source industries.

2. Extraction, removal or recovery

Natural gas which contains varying amounts of CO\textsubscript{2} and H\textsubscript{2}S is called sour natural gas. Hydrodesulphurization or natural gas sweetening is a process for H\textsubscript{2}S removal from natural gas. Hydrodesulphurization of natural gas with subsequent sulphur recovery is mainly performed in amine and Claus plants. For as for large quantities of sulphur (> 50 ton/day), these processes are most economical. However, in case of H\textsubscript{2}S removal from smaller gas fields, the gas is generally treated by liquid redox processes, or, alternatively, by amine treatment followed by incineration or re-injection of the acid gas in an empty well. There are some new technologies in the filed as well. All these processes have their strengths and weaknesses \cite{6}.

2.1. The Frasch process

The Frasch process developed in the 1890’s by chemist Herman Frasch. Sulphur is extracted from underground deposits by the Frasch process until the end of the 20\textsuperscript{th} century. It is carried out by putting three concentric tubes into the sulphur deposit. Then, superheated water and hot air are put into the tubes, so the sulphur is pushed up to the surface. This kind of recovered sulphur is usually very pure, but if contaminated by organic compounds there is no need to purify it.

2.2. Amine process

The most common gas treating process is the amine process in which acid gases react chemically. The 'rich' amine solution is heated under low pressure to regenerate the liquid by forcing off the acid gases. In this process, several different amine solutions can be used such as: methyl di-ethanol amine (MDEA), mono-ethanol amine (MEA) and di-ethanol amine (DEA). For each of the amine solutions listed, the sweetening process is similar. The type of solution used in the process depends upon the type and quantity of acid gas contained in the sour gas stream and the volume of sour gas to be treated \cite{7}.

2.3. The Claus process

Nowadays, the standard process used to extract sulphur via petroleum and gas sources is the Claus process developed in the 1880’s by chemist Carl Friedrich Claus. For quantities of sulphur greater than 50 ton/day, this process is considered economical. In this kind of sulphur needs to be processed to remove it from the natural gasses that are mixed with it. In the Claus processing, the sulphur compounds in the gas are converted into elemental sulphur, and the hydrogen sulphur is extracted from the gas. Sulphur recovered this way may be in a solid or a liquid form.

Hauwert \cite{8} reviewed four processes for small-scale sulphur recovery from sour associated gas, including liquid reduction/oxidation (redox), Thiopaq O&G, CrystaSulf, and direct catalytic oxidation. He described the processes for removal of sulphur quantities ranging from 1 ton per day to 20 tons per day, for direct treatment of high-pressure natural gas and low-pressure associated petroleum gas, and for sulphur recovery from low-pressure acid gas.

2.4. Liquid redox

The liquid redox processes are suitable for recovery when the sulphur load is too small for the Claus process. In Redox methods an alkaline solution with high-valent metal ions is used, such as vanadium (Stretford process) and iron (LoCat and SulFerox processes). The metal
ions convert the dissolved H$_2$S into elemental sulphur. Then, it is regenerated by oxidation with air. The sulphur content is generally 80% by weight (dry) and can be upgraded to 99.5% by washing and smelting. Plugging and foaming may occur and cause operational problems due to the process solution is aqueous with hydrophobic (i.e., repel water) solid sulphur particles [8].

### 2.5. Thiopaq O&G

Thiopaq O&G process, developed in the 1990s by Paques for desulphurization of biogas, was modified by collaboration with Shell Global Solutions for application at high pressure in oil and gas environments. The first commercial Thiopaq O&G unit was built in 2002. It is a biotechnological process which removes H$_2$S from gaseous streams by absorption into a mild alkaline solution followed by the oxidation of the absorbed sulfide to elemental sulphur by naturally occurring microorganisms. Offers a replacement for liquid redox processes or amine treating, Claus recovery and tail gas treatment; minimal chemical consumption, high turndown ratio, gas treatment as well as sulphur recovery, H$_2$S removal to below 4 ppmv, essentially 100% conversion of sulfide in the bioreactor with 95-98% selectivity to S, no need to replacement of the biocatalyst. Application to H$_2$S concentrations of 100 ppmv to 100 vol.% and pressures from 1-75 bar(g). Thus, direct treatment of either the sour gas or the amine off-gas is possible [6]. This process is shown in Figure. 1.

![Fig. 1. The Thiopaq O&G desulfurization process uses naturally occurring Thiobacillus bacteria to oxidize hydrogen sulfide to elemental sulfur. Source: Paqell](image)

In this processing method, in comparison to traditional removal processes such as the Amine/Claus process the following advantages are recognized: no practically free H$_2$S available anywhere downstream of the scrubber inlet leading to very safe and easy to operate unit; very simple line-up requiring little control and supervision; no complex control loops; a relatively large volume of (cheap) solvent leading to very slow changes in solvent composition and performance of the unit and more robust process [6].

### 2.6. CrystaSulf

In the 1990s, The Gas Research Institute developed the process for H$_2$S removal from high-pressure gas in which a non-aqueous hydrocarbon solvent which contains sulphur dioxide is used similarly to a liquid-phase Claus process . When the solution comes into contact with the gas containing the H$_2$S, elemental sulphur and water are formed, the sulphur dissolves in the solvent and is precipitated in a scraped-surface crystallizer to decrease plugging and foaming. The amount of H$_2$S is reduced to ppmv levels, and for associated gas applications, no foaming or negative effects from hydrocarbon condensation are expected. However, the solvent can be costly for large, low-pressure gas flows with limited H$_2$S content [8]. Figure 2 illustrates the CrystaSulf process.
2.7. Direct catalytic oxidation

This process was developed by TDA Research, which is licensed by GTC Technology. The process (Figure 3) which is applicable to lean acid gas streams, converts \( \text{H}_2\text{S} \) directly into sulphur by using a catalyst. As the company said, it is effective for sulphur recovery ranging from 0.1 ton per day to 200 tons per day and that the sulphur conversion efficiency is approximately 90% in a single pass [8].

3. Application

There was a remarkable investment in clean air and against discharge of sulphur into the atmosphere during the 1960s. This made sulphur a surplus commodity on the market, particularly in the United States and Canada. Therefore, the interest appeared regarding the application of sulphur in different fields, and extensive research programs initiated with the focus on various properties of the material.

Most elemental sulphur is converted into sulphuric acid (\( \text{H}_2\text{SO}_4 \)) which is used in the industrial production of chemicals and in the sulphur-iodine cycle to obtain hydrogen. The largest use of sulphuric acid is for the manufacture of primary phosphates, nitrogen, potassium, and sulphate fertilizers. Sulphuric acid often used to manufacture numerous products including industry chemicals, construction materials, paints, rubber products, medicines, fibers, sugar, plastics, paper, lead-acid batteries, non-ferrous metals, pigments, hydrofluoric acid, carbon disulphide, water treatment, caoutchouc, dyestuffs, drilling muds, in petrochemical and pulp and steel pickling, fungicides, pesticides, and pharmaceuticals.
3.1. Sulphur in agriculture: fertilizer

For many years, the focus of fertilizer industry has been on nitrogen, phosphorus, and potassium; however, that attention is expanding to include plant nutrient sulphur as sulphate. Sulphur is typically considered the fourth major nutrient in terms of total volume requirements for plant growth [9]. Sulphur is necessary for plant growth and nutrition. It contributes to an increase in crop yields in different ways: it provides a direct nutritive value; it provides indirect nutritive value as soil amendments; and it improves the use efficiency of other essential plant nutrients, particularly nitrogen and phosphorus.

Sulphur contained fertilizers are of two primary kinds; sulfate fertilizers where the sulphur is in sulfate form and readily available for crops to intake, and elemental sulphur fertilizers which need to be oxidized into sulfate before plants are able to uptake it [10].

The traditional fertilizers containing sulphate includes single superphosphate, ammonium sulphate, and potassium sulphate. The emerging fertilizers containing sulphate are ammonium phosphate-sulphate, potassium magnesium sulphate, various micronutrient sulphate salts, ammonium nitrate-sulphate, sulphate-NPK compound fertilizers and urea-ammonium sulphate [11].

The sulphur formed from the THIOPAQ desulphurization plant is ideal for use as a fertilizer due to having a small particle size which helps in the oxidation of sulphur into sulfate, the effluent sludge from the THIOPAQ process contains sodium salts, sulfate and carbonate, which are all useful nutrients [12].

Worldwide sulphur deficiencies (Figure 4) are increasing due to a reduction in traditional S-containing materials, cleaner air programs and intensified agriculture increasing crop demand which leads Sulphur to gain prominence as a major nutrient [11]. It also provides an opportunity to advance carbon sequestration through the removal of a primary constraint to biomass production.

![Figure 4](source: TSI)

3.2. Sulphur in construction

In recent years, sulphur is increasingly used in road construction because the most significant consumers of sulphur surplus can be construction and road-building industries. Construction materials such as sulphur concrete and sulphur asphalt is receiving more attention since they are environmentally friendly and cost-effective. Many research [12-13] proved that in order to obtain composite resistant to chemical aggression, the sulphur could be used as a bond in the sulphur concrete. granular form is preferred for this area because it is more technological and creates no dust.

3.2.1. Sulphur concrete

Sulfur concretes is the mixtures of mineral aggregate and modified sulfur (sulphur-polymer) as a binder. The study of sulphur concrete began in the 70-ies of the 20th century, and its first application was for building ship anchors. In 1921, Bakon and Davis [14] introduced sulphur properties and its application in the production of construction materials.
The primarily produced sulphur concretes had several disadvantages - low resistance to high temperatures (this drawback has remained till now, and it is caused by the fact that sulphur melting temperature is 120°C), low fire resistance and cracking during hardening of large sulphur concrete volumes. Technology development allowed to eliminate the majority of the drawbacks. Thus it was determined that addition of plasticisers (especially polysulfides) to sulphur binders would not only improve plasticity of the solution but also decrease cracking, while the addition of stabilizers including dicyclopentadiene would increase fire and atmospheric conditions resistance of sulphur concrete. Moreover, the addition of Fire-retardants and Antiseptics decrease sulphur composition combustibility and increase the biological stability of sulphur concretes, respectively [9]. Figure 6 illustrates the technological scheme for sulphur concrete manufactures production.


Sulphur concrete has many advantages such as: frost resistance, high strength, stability in corrosive medium, low water permeability, high corrosion resistance, fast strength development, utilisation possibility of most of the harmful substances including radioactive wastes, and recycling possibility [15]. One of the key advantages of sulphur concrete is the little time it takes to harden that propitiates a kind of application in which it can offer a great advantage in repairing rigid (concrete) pavements, and urgent repairs in big industries [16]. In Table 1. the comparative characteristics of sulphur concrete and Portland cement concrete is presented [9].

Table 1. Comparative characteristics of sulphur concrete and Portland cement concrete

<table>
<thead>
<tr>
<th>Properties</th>
<th>Sulphur concrete</th>
<th>Concrete</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture resistance</td>
<td>1</td>
<td>0.8</td>
</tr>
<tr>
<td>Chemical resistance (to acids)</td>
<td>84%</td>
<td>23%</td>
</tr>
<tr>
<td>Frost resistance (at 100% humidity)</td>
<td>300</td>
<td>50</td>
</tr>
<tr>
<td>Attrition resistance, %</td>
<td>3%</td>
<td>17%</td>
</tr>
<tr>
<td>Compression strength, MPa</td>
<td>55-65</td>
<td>15-25</td>
</tr>
<tr>
<td>Bending strength, MPa</td>
<td>10-15</td>
<td>6-9</td>
</tr>
<tr>
<td>Tensile strength, MPa</td>
<td>5-7</td>
<td>3-4</td>
</tr>
<tr>
<td>Time of strength development, hr</td>
<td>0.3</td>
<td>24</td>
</tr>
</tbody>
</table>
Utilization of sulphur-polymer materials is most efficient in some fields including: 1) Chemical industry: building vessels for chemically active substances (including chemical wastes), foundations for chemical vessels, concrete floors for chemical facilities, containers for chemical product storage, etc.; 2) Hydrotechnic construction building: manufacturing pipes for gravity and pressure sewer systems; pipelines for corrosive and toxic waste waters; facing slabs for canals; collector rings; drainage pipes; 3) Hazard waste disposal: Sulphur polymer materials allow to solve a problem of disposal of solid toxic wastes (mercury-containing wastes, radioactive wastes, overdue medical substances, dry pesticides) [9].

### 3.2.2. Sulphur asphalt

Bitumen, which is hard to obtain as a raw material, can be replaced by sulphur which is an abundant and practical raw material. Sulphur asphalt (SA) which is sometimes called sulphur bitumen or sulphur extended asphalt (SEA), is an alternative for asphalt road binder, a process in which sulphur is used to extend asphalt materials as a means of energy conservation by minimizing asphalt demand [5].

Commercial development of sulphur asphalt road paving materials started during the oil embargo in the 1970s. Early attempts in sulphur enhanced asphalt showed positive improvements in the asphalt properties; however, it suffered from the formed H$_2$S during the manufacturing of the asphalt. New interest in using sulphur as a construction material has led to the development of sulphur enhanced asphalt with much better quality than standard asphalt [17]. Sulphur asphalt can create additional markets for the sulphur surplus and extend the overall asphalt products market.

### 3.3. Lithium/sulfur batteries

Recent decades witness worldwide attention towards new energy sources due to the impact of CO$_2$ emissions on global warming effect. In this regards, electrochemical energy storage systems, and most particularly batteries, play a crucial role in current and next generation applications. One such promising technology is lithium/sulfur (Li/S) systems with the potential for extremely high gravimetric energy [18]. The interest in Li-S is driven by the advantages it has over the existing Li-ion technologies. Many research teams are focused nowadays on the development of Li/S technology, fundamental understanding and modeling, and application-based control algorithm development [19], and at present, there are two start-up companies-Sion Power [20] in the USA and OXIS Energy [21] in the UK – on the market.

The Li-S battery’s working mechanism is complex; however, Fotouhi [19] summarizes the working principle as follows.

“In a fully charged state, a Li-S cell consists of a cathode usually containing sulfur, a carbon-based material and a binder. The anode is lithium metal and is separated from the cathode by a polymer separator and an organic-solvent based electrolyte. During operation (discharge), solid sulfur from the cathode dissolves into the electrolyte, forming S$_8$(l). Liquid S$_8$ is then electrochemically reduced at the cathode to form intermediate products, so called lithium polysulfide species (Li$_2$S$_x$) with accompanying oxidation of Li metal to Li$^+$ ions at the anode. The polysulfides species (Li$_2$S$_x$, 2 < x < 8) are soluble in the liquid electrolyte and diffuse out from the cathode to the electrolyte/separating side. When the discharge proceeds, the length of the polysulfide chain is getting reduced, which in turns affects the viscosity, mobility and solubility of Li$_2$S$_x$ compounds. At the end of discharge, S$_8$ is fully reduced to S$^{2-}$ (Li$_2$S), and the anode is fully stripped of Li metal. This process is demonstrated schematically in Figure 1. During charge, the reverse reactions occur, with Li$^+$ ions depositing at the anode as Li metal and low-order polysulfides oxidizing from S$^{2-}$ up to S$_{8-2}^2$ and eventually S$_8$(s).”

By developing state of the art in modeling, it is envisaged that it will be possible to explore state-of-the-art techniques for rapid online execution of spatially distributed models [22]. Auger [23] represents a possible road map (Figure 6), including contributions by control engineers follow developments in fast-executing electrochemical models.
3.3.1. Advantages and limitations

One of the key advantages of the Li-S battery is the higher specific energy, predicted to be 2–3 times higher than the excited li-ion batteries. Another advantage is that sulphur is cheap, abundant and non-toxic and environmentally friendly. Other advantages of Li–S batteries include low cost and availability of sulfur; intrinsic protection mechanism from overcharge, providing safety; wide temperature range of operation; possibility of long cycling [19-24]. In addition, the Li–S battery can be operated over a wide temperature range, especially at low temperature, which is a good power source in cold environments, such as battery systems for electric vehicles, as well as some aeronautical applications [25].

Despite the advantages of the Li-S battery technology, it suffers from some limitations as well. The complex reaction mechanism involved in the conversion of elemental sulfur ($S_8$) to the final reduction product, lithium sulfide ($Li_2S$) is a major difficulty in Li-S materials and cell development [22]. It also suffers from limitations such relatively low practical specific energy (200–300 W h/kg) against expected values of 450–650 W h/kg; rapid decrease in capacity during cycling (0.1–0.4% per cycle); high self-discharge rates, high self-discharge and short cycle life, particularly in the presence of high discharge currents [19-24]. This problem arises from the following reasons: insulating nature of sulphur and Li-S; dissolution of lithium polysulfide intermediates that occur during battery charge and discharge; large volume change during intercalation processes due to a reduction from elemental sulphur to Li-S; use of Li metal electrode; shuttle effect and self-discharge [26].

3.3.2. Application

Current applications of Li-S battery technology is limited to low power and cycle life requirements such as high altitude long endurance unmanned aerial vehicles (HALE UAVs). Future applications of the Li-S battery were then discussed in three areas, the need for more cycle life, the need for more power, and the need for both [27].

4. World production and consumption

Over the last two decades, sulphur production has changed from a mining industry to almost a co-product of the oil and gas industry. Over 25% of elemental sulphur all over the world originates from the desulphurization of fossil fuels, and alone in Europe, it is already 38.6%. Elemental sulphur, accounts for over 97% of world elemental sulphur output, while mined
sulphur is in retreat. Production via the Frasch process ceased in the United States in 2000 and is in decline elsewhere [9]. In 2017, the world production was 83 000 m/t, and the largest producers were China, USA, Russia, UAE, Canada and Saudi Arabia, respectively [28].

Fig. 7. World Sulphur Production in 2016 and 2017. Source: OEC

In the developed world, much of the additional sulphur from clean fuels has already occurred; however, potential production from the developing world due to stricter regulations limiting sulphur content looms large. A large source of sulphur output is regulations restricting sulphur content in marine bunker fuels [9]. In 2016, the International Maritime Organization (IMO) has set a global limit for sulphur in fuel oil used on board ships of 0.50% m/m (mass by mass) from 1 January 2020. The current global limit for the sulphur content of ships’ fuel oil is 3.50% m/m (mass by mass). That means refineries will have to be configured to strip out excess sulphur [29].

Over the last decade, sulphur consumption has also increased in the world. China is the leading consumer with demand across fertilizer and other sectors representing over a quarter of global consumption. However, its attempt in development of domestic production will reduce China’s dependence on foreign supply. In 2017, the largest consumers were China, Cuba, Australia, Chile, Sought Africa, Congo, respectively [28].

In terms of lime sulphur demand, the largest and fastest growing region for lime sulphur is the Asia Pacific. North America is also anticipated to boost the market growth of lime sulphur in the market. Other regions expected to boost market demand for lime sulphur are Europe, Latin America and Middle East & Africa during the forecast period [4].

5. Balance, logistics and outlook

Increased supply leads to elemental sulphur inventories rise, and prospective surpluses (figure 7) will further add to it and stocks are expected to play increasing roles in world sulphur markets. Increased development of sour gas reserves and refining of sour crudes combined with growing government restrictions requiring sulphur removal from fuels leads to sulphur outputs. Thus, the sector will face serious unbalance problems in the near future unless consideration is given to the increased supply streams [9]. At the end of 2016, some experts [30] reported that global market is facing oversupply and forecasted it continues till 2020.
The main functions of sulphur logistic concept include: procurement, production, distribution and disposal. The changing nature and dynamics of sulphur production and consumption always are having great effects on sulphur logistics worldwide. The non-voluntary nature of today’s production coupled with still cyclical demand industries has interfered with logistical sulphur structures, particularly on the production side. With the implementation of IMO’s 0.5 per cent sulphur cap, the costs and logistics of compliance are at the forefront of discussions for professionals across the industry. It is necessary that operators and suppliers work together to ensure fuel availability past the cutoff point and the shipping industry will have to consider a switch to alternative fuels, such as marine gas oil (MGO), or install scrubbers, a system that removes sulphur from exhaust gas emitted by bunkers.

According to Heffer and Prud’homme, a new supply of exportable sulphur in 2016 in West Asia and EECA Global sulphur production will grow by 4% p.a. compared with 2015, reaching 72 Mt S in 2020. during the next five years, the largest increases in production will occur in the sulphur exporting regions of West Asia and EECA, each at 6% p.a. Moderate growth of global sulphur demand will occur in the near term, but the current balance is shifting to potential surpluses. Global consumption of elemental sulphur will grow at an annual rate of 3% compared with 2015, reaching 69 Mt S in 2020 and the global supply/demand situation will shift from a near equilibrium condition in 2015 to the emergence of growing surplus towards 2020.

6. Summary

In the last decades, sulfur availability has considerably grown in many countries as a by-product of the petroleum industry, mainly due to the current environmental restrictions regarding the petroleum and gas refining processes, which limit the maximum quantity of sulfur present at combustibles. In the past oil and gas companies were primarily by-product suppliers, they are now the primary producers.

Sulphur has applications in different fields, the most important of which are agriculture, construction, and energy source industries. Sulphur is becoming an important fertilizer nutrient, with soil deficiencies reported in many regions of the world. Sulphur consumption for industrial uses is likewise showing some exciting changes. sulphur asphalt and sulphur concrete seem to be more employed as construction materials. Electrochemical energy storage systems, and most particularly batteries, play a crucial role in current and next generation applications. Thus, lithium/sulfur (Li-S) systems are one such promising technology with the potential for extremely high gravimetric energy.
Over the last two decades, sulphur production has changed from a mining industry to almost a co-product of the oil and gas industry, and sulphur production and consumption have also increased in the world. In 2017, the largest producers were China, USA, Russia, United Arab Emirates, Canada and Saudi Arabia, respectively. The largest consumers of sulphur are China, Cuba, Australia, Chile, Sought Africa, Congo, respectively.

The development of necessary and flexible logistical chains to match evolving production and consumption is very important to the sulphur marketplace. These issues need to be addressed by producers and consumers to remove uncertainty, discontinuity and possible refinery shutdowns.

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