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Original Research

Recent Developments in High Sulfur-Content Polymers

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Abstract:

Synthetic polymers are one of the most widely used materials, and there is a growing demand to produce new polymers that are more environmentally friendly. Despite the fact that sulfur, a petroleum by-product, is viewed in polymers' manufacture as a potential replacement for carbon, it cannot form a stable polymer alone. But it responds to combining sulfur polymers with organic crosslinking molecules in a process known as "inverse vulcanization," involving high temperatures and extended reaction periods, so that they are stable and resist decomposition. We will highlight recent advances in sulfur (S8) polymerization for the production of high sulfur polymers, as well as future prospects. Although homo polymerization of S8 has been known for a long period of time, this kind of polymeric sulfur is chemically unstable and reverts to its original state. In this article, we shall explore the history of sulfur production as well as the limitations of using sulfur as a chemical reagent in the synthesis of novel materials and composites. S8 was pioneered as a reaction medium and co-monomer in inverse vulcanization in order to develop chemically stable and process able sulfur copolymers that were also combustible.

Keywords: *sulfur, elemental Sulfur inverse vulcanization, Li-S batteries, sulfur polymers*

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1. Introductions:

Polymers have been closely associated with life in the present world since the beginning of existence (e.g., cellulose, starch, and natural rubber). Synthetic polymer materials have been researched since the middle of the nineteenth century. The polymer industry has rapidly developed and has become larger than copper, steel, aluminum and a few other commodities in the industry [2]. Useful resources and alternative feedstocks are needed to enable the long-term production of polymers. The topic is examined in the context of a novel family of sulfur-based polymers [3]. Sulfur is known since time began and exists uncombined in nature. It is an important global pollutant when oxidized to sulfur dioxide. Sulfur compounds are often used in the current industrialized environment. Since the expenses of using the industry's sulfur fuel gas, oil and solid fuel supplies are so high, both feedstocks and refining effluent must be cleaned up to reduce production costs and halt contamination. [4]. Seven million tons of excess sulfur are being produced each year, according to the most recent estimates. Elemental Sulphur is a byproduct of production and is held in massive storage facilities that have a negative impact on the environment and human health [21-22]. A list of actions on the environment has been produced, including strategies for reduction of SO_2 emissions. Further steps were implemented, such as "cleaner" energy resource usage campaigns, recycling of waste and conservation of natural resources. In novel chemical synthesis sulphuric acid is used as a reagent and is also used for the chemical and petrochemical industries as a catalyst. As a result, huge quantities of sulfuric acid waste are produced, containing organic pollutants. The usage of sulfuric acid waste has a considerable environmental effect and can be recycled in several techniques [5].

Elemental sulfur production from oil refineries and gas purification plants provides for about two-thirds of total sulphur production [20]. As a result of their sulfur content, sulfur-containing polymers [23], including the sulfur-random-styrene (SRS) group, exhibit distinctive electrical, absorption, and optical characteristics. Aside from the fact that they are resistant to heat and chemical assault as well as radiation, they are also biocompatible and have a high affinity for heavy metal ions [1].

2. Methodology:

Documentary, library, and scientific article methods were used in this study to write a review about high sulfur content polymers, which have been getting a lot of attention recently because of their possible low cost and wide range of applications.

3. Rechargeable metal–sulfur batteries

Lithium-sulfur (Li-S) [19] batteries have become one of the most widely utilized low-cost energy storage systems in recent years. The lithium poly sulfides (Li_2Sn, whereby n > 4) used in Li-S batteries are an electrical and ionic insulator, as well as a weak sensitive material that creates soluble species during the demobilization process [6]. When compared to today's most advanced lithium-ion batteries, lithium-sulfur batteries are guaranteed to retain four to five times the amount of energy stored by the latter kind of battery. According to the findings of the researchers, this kind of polymer has the potential to be the answer for lightweight, cost-effective batteries that can store large amounts of energy. Inverse vulcanization was used to produce sulfur-rich copolymers based on poly(sulfur-random-1,3-di-isopropenyl benzene) (poly(S-r-DIB)) that were used as cathode materials for lithium-ion batteries [16].

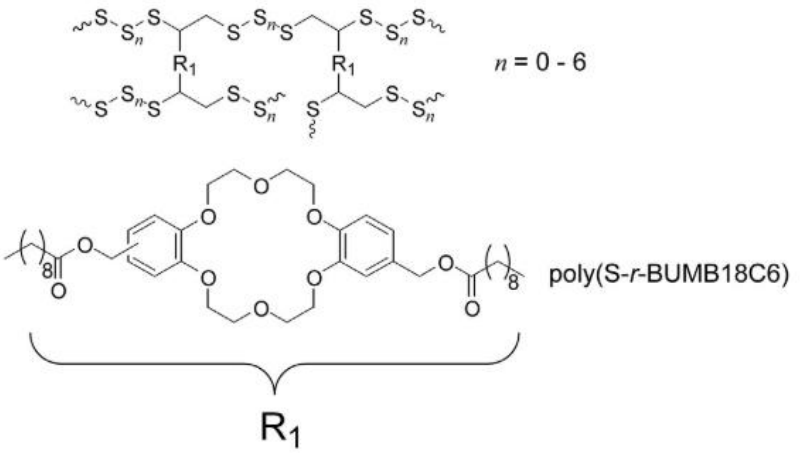
A sulfur composite cathode, an organic electrolyte, and a lithium metal anode are required to build a LiS cell [28].This means that when the battery is joined together, it is in the charged condition. Both sulfur and lithium may be found in their neutral forms in the environment. In this case, when the battery is completely depleted, the lithium metal is oxidized at the negative electrode (anode), resulting in the formation of lithium ions and electrons. Throughout the liquid electrolyte, the lithium ions migrate from the negative electrode to the positive electrode, and vice versa. The electrons pass across an external electrical circuit, causing an electric current to flow through the circuit. During the process, sulfur mixes with lithium ions and electrons at the positive electrode (cathode) and is reduced in a series of complex processes to form lithium sulfide, which is the product that is produced at the conclusion of the process (Li2S)[29]. When the battery is re-charged, the back responses begin to occur once again. The lithium ions and electrons from the positive electrode are removed, allowing sulfur and lithium metal to be re-formed at the cathode and anode in their respective positions [30].

In this study, we also attempt to describe the covalent adjustment hypothesis, which is based on new research on covalent sulfur-containing composites in a variety of rechargeable metal–sulfur batteries [12]. The most serious issues that need to be addressed with this technology are the quick capacity loss and low cycle lifetimes of Li-S cells. It has been shown that two factors are principally to blame for these difficulties: dissolution of the active sulfur material from inside the cathode and mechanical degradation of the composite cathode structure [31].

4. Copolymerization of elemental sulfur

The traditional poly(s-bis(alkenyl)compound-a) synthesis technique is as devised for testing of the magnetic sulfur and bis(alkenyl) compound (BUMB18C6) tube equipped with a magnetic stirring rod, then heated up to 159,85 ºC for ten minutes in a temperature-controlled oil bath. The mixture was kept at 159,85°C in the oil bath for a period of 1 to 1.5 hours after the stirring was completed. In less than a minute, a spatula whisked it from the test tube to room temperature. This is a polyurethane product (S-r-BUMB18C6-0.5) product. The 0.776 g of poly (S-r-

BUMB18C6-0.5) (yielding about 100 percent) was summarized. A poly(S-r-BUMB18C6–0.1) was synthesized with a high yield using a sulfur and BUMB18C6 mixture (Scheme 1.). In order to produce the bis(alkenyl) using the crown ether, the elemental sulfur was used to make the poly (S-r- BUMB18C6-a)(a 14%, 0.1 or 0.5), a soluble and elastic copolymer that was produced from the crown ether. Poly(S-r-BUMB18C6-0.5 is the polysulfide that has been proven to be electrochemically active. The performance of the test cell Li poly(S-r-BUMB18C6-0.5) was excellent under continuous charging-discharge conditions, and it was also suitable for other applications [7].

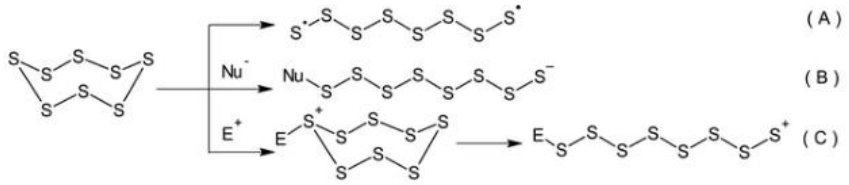


Scheme 1. Structure of a copolymer combination that has been reported (S-r-BUMB18C6-0.5)

5. Inverse vulcanization

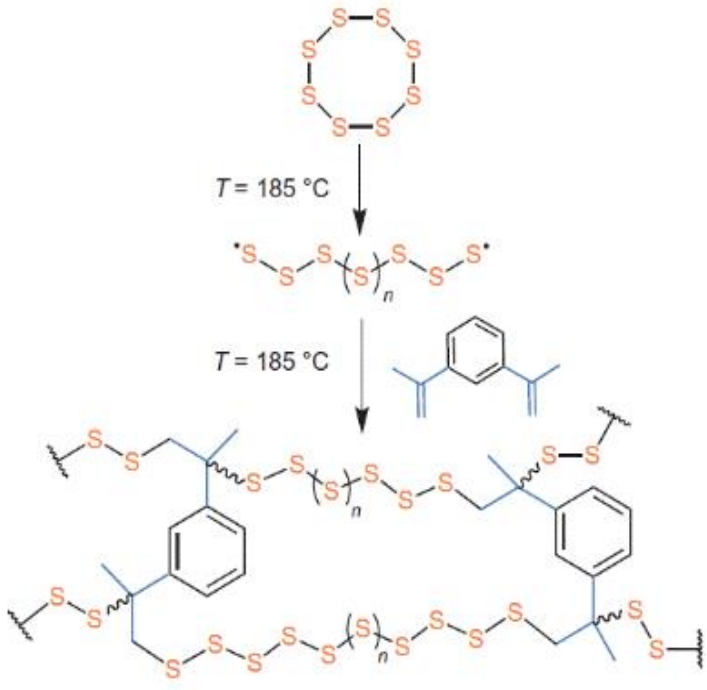
A method called "inverse vulcanization" can be used with any two-, three-, or more-vinyl, or alkenyls *co*-monomer that can be mixed with liquid sulfur and that can undergo thiol-ene/thiol-yne reactions [25-26].

Given the all-encompassing availability of sulfur, the use of it for polymers and substances was revitalized. Inverted, high sulfur polymers in vulcanized applications [27] have recently been highly attracted by their possible low cost and many uses. The hydro desulfurization of crude oil and gas supplies more than 60 million tons of surplus sulfur per year [8]. Depending on the circumstances, the opening of the S8 ring may be mediated by a radical or an ionic process. The latter technique allows for catalytic sulfur activation, either nucleophilic or electrophilic (scheme 2.) [24].



Scheme 2. The processes of opening the S8 ring are radical (A), nucleophilic (B), and electrophilic (C).

According to current research, a sulfur polymer generated by reverse vulcanization has recently been shown to have better process ability and mechanical qualities under ambient conditions [13]. For the first time, Pyun and colleagues described "inverse vulcanization," which increases the durability of sulfur polymers by using a small organic molecule that acts as a cross-linker to depolymerize the polymer (Scheme 3.), such as 1,3-di isopropenyl benzene (DIB), to prevent depolymerization. It is possible to produce thermodynamically stable polymeric glasses by immediately bulk copolymerizing liquid sulfur with 1,3-diisopropenyl benzene (DIB) when the temperature is raised to 185 °C by inverse vulcanization. Using DIB as a catalyst, the inverse vulcanization of S8 resulted in the formation of high sulfur content polymers that were electrochemically active and may be employed as the electroactive material in lithium-ion battery cathodes, according to the researchers [17].



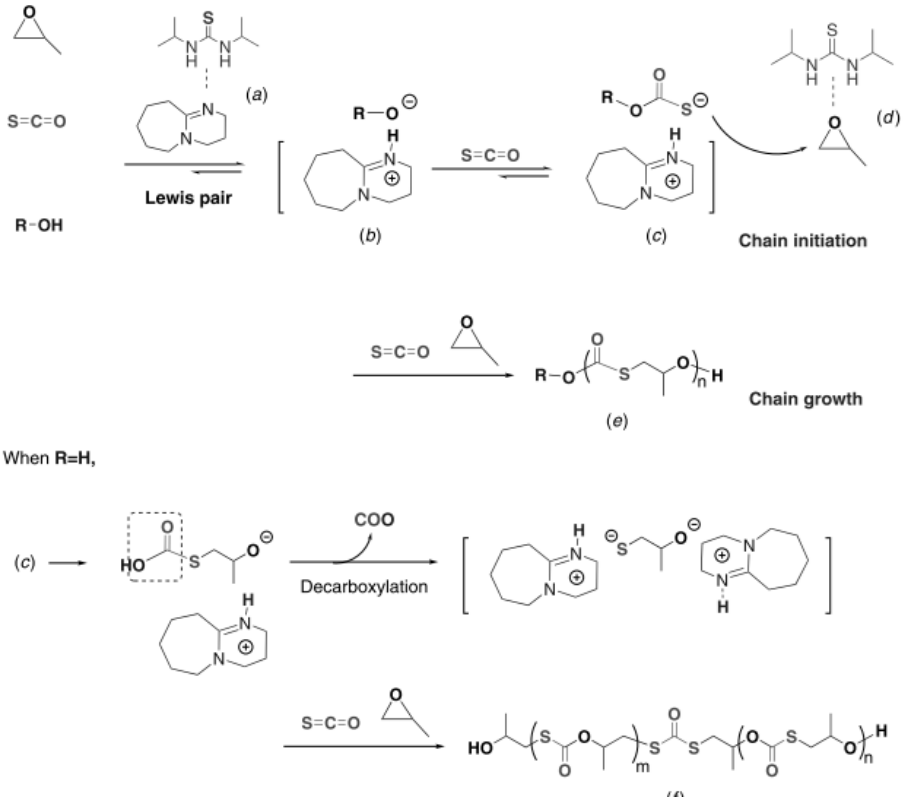
Scheme 3. The production of poly(S-*r*-DIB) co-polymer using DIB cross-linker

Sulfur base polymers may be synthesized in an ecologically acceptable manner using a catalytic inverse vulcanization technique. This approach works with a wide range of catalysts, including metals that are both inexpensive and non-toxic. Reverse vulcanizations without catalysis improve polymer characteristics while generating less toxic H_2S gas than traditional vulcanization processes do. They also reduce reaction temperature and duration. With these characteristics, it is quite likely that we will be able to produce and utilize these extraordinary and rare materials.

6. Sulfur-containing polymer matrix synthesized using dual organ catalysts

Cooperative catalysis can be done when organic Lewis pairs are used as hydrogen-bond donors and bases (e.g., phosphazene and guanidine). It has been shown that metal-free live copolymerization of carbonyl sulfide with epoxides can be done. An active copolymerization of COS with multiple epoxides with high activity, using ordinarily prepared thioureas (TUs) and organic LBs (8-diazabicyclo[5.4.0]undec-7-ene(DBU); N-methyl-1,5,7-triazabicyclododecene(MTBD);1-tert-butyl-4,4,4-tris(dimethylamino)-2,2-bis [tris(dimethylamino)- phosphoranylidenamino]-2λ5,4λ5-catenadi(phosphazene) (^{t}Bu-P4); 1-tert- Butyl-2,2,4,4,4-pentakis(dimethylamino)-2λ5,4λ5-catenadi(phosphazene)(^{t}Bu-P2); and tert-butylamine-tris(dimethylamino) phosphorene)(^{t}Bu-P1)) has been developed. It was important to comprehend the catalytic process of joining Lewis pairs consisting of TU and base, as well as a non-covalent way to activate and stabilize the alcohol initiator/chain end for controlling the anionic copolymerization, in order to optimize the performance of this catalyst system. Poly (monothiocarbonates) that are made are transparent and have a correct composition, controlled molecular weights, and narrow PDIs.

Poly(monothiocarbonate) has previously been synthesized from COS and organic bases under mild conditions using thiourea and organic bases (Scheme 4.). Thioureas and BnOH, on the other hand, were shown to be more effective than other systems employed in this work in terms of COS/epoxide copolymerization as well as catalytic activity and selectivity. These metal-free catalyst systems have a major benefit in that they may be employed with a range of epoxy compounds without losing their potency. An effective strategy for producing metal-free, sulfur-containing polymers with outstanding activity and selectivity might be achieved via the employment of these organic thiourea and base thiols and bases [9].



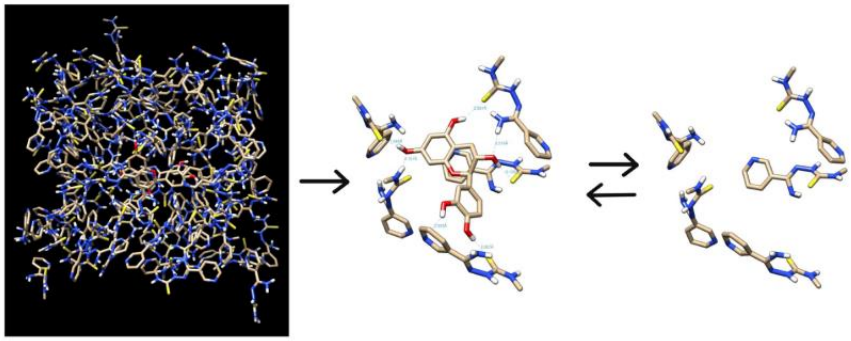
Scheme 4. A proposed mechanism for anionic copolymerization in the presence of BnOH and H2O, the TU-1/DBU pair, catalyzes the production of a chain.

7. Sulfur molecularly imprinted polymers

Researchers have discovered novel sulfur-based monomers with anticancer and antioxidant properties after comparing their computational similarities to conventional laboratory approaches. Molecularly imprinted polymers (MIPs) have been demonstrated to be influenced by the antioxidant capabilities of polyphenol molecules found in catechin, which serve as a template molecule for the polymerization of other molecules. This happened in the middle of a virtual box with certain dimensions, which was then charged with the largest amount of monomers possible in order to make it look like the real thing. Finally, this box is subjected to four minimization procedures in order to reset its molecules in order to get the least energy set, assuming that the first mixing phase in actual pre-polymerization mixture generation is taken into consideration. After the box had gone through a binary equilibration phase, the least amount of energy was sent out

of the box. After reaching 333 K at atmospheric pressure in the first stage of equilibration, it is required to maintain that temperature for the duration of the second step in order to mimic the temperature variations that occur during real polymerization. This is a two-part process [10].

Before the polymerization process started, a portion of porogen was inserted in a 20 mL flask and the polymerization process began. A thiosemicarbazone-based ligand ($C_6H_8N_6S$, $C_7H_{10}N_6S$, $C_8H_{11}N_5S$, $C_8H_8N_6OS$, $C_8H_9N_3OS$ and $C_{14}H_{17}N_5O$ for ligands 1–6), ethylene glycol di-methacrylate (EGDMA) was added to dilute the template (Scheme 5.). The quantities of template and monomer were then modified depending on the simulation findings. Finally, a nitrogen stream was used to complete the polymerization process, which was then followed by a 24-hour soak in warm water at 60 ^oC [11].

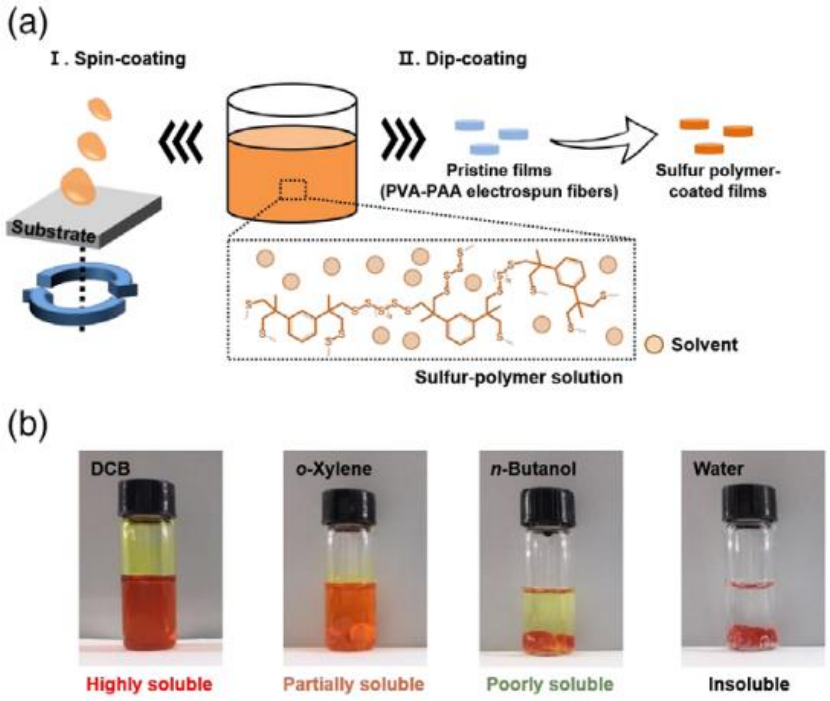


Scheme 5. MIP computational simulation Specific cavities for ligand 3 – catechin MIP.

8. Nano fibrillary Films with Sulfur-Polymer Coating

During radical polymerization, diene–dithiol is a low-cost, functional, renewable monomer that may be used as a building block. The researcher has come up with a solution-based way to make thin films of sulfur polymers by spin-coating or dip-coating. When elemental sulfur, 1,3-di-isopropenyl benzene, and other organic solvents are mixed together, a sulfur polymer is formed. The solubility of the sulfur polymer in different organic solvents has been investigated [13]. A simple dip-coating process is used to coat the electrospun (PVA–PAA) nanofiber film with a sulfur polymer layer (100 nm) that is both efficient and uncomplicated (Scheme 6.)[15]. By using the sulfur polymer-coated film as an output for mercury ion extraction, it is possible to show that sulfur may be produced on multiple materials surfaces while still retaining its elemental sulfur properties [14]. Using DCB's sulfur polymer solution, it has been shown that sulfur polymer can be applied evenly to scaffolds with high surface energy (> 40 mN/m). The sulfur polymer was created by immersing a sealed container containing sulfur and diisopropyl benzene (DIB) in a bath of oil at a temperature of 0 degrees Celsius for 6 to 7 minutes. Other solvents

included n-butanol, 1,2-dichlorobenzene, o-xylene, and deionized water for dissolving sulfur polymer. Sulfur polymer and solvent were heated in an oil bath for 12 hours to a temperature close to the boiling point of the solvent. This reaction was done with each sealed container for 12 hours [13].

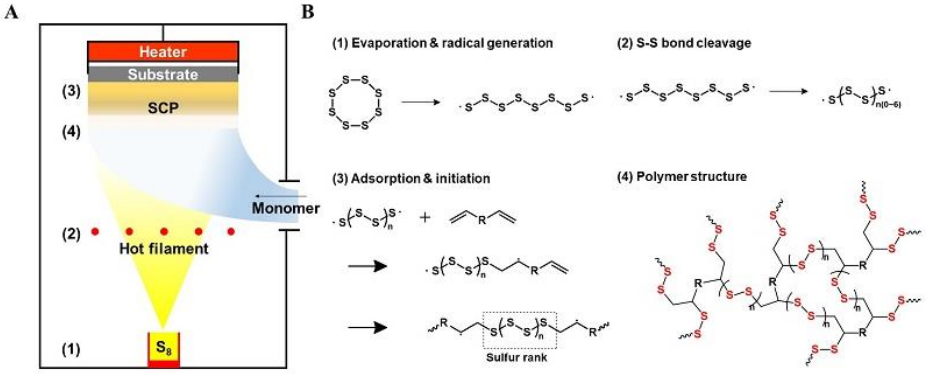


Scheme 6. Solubility test of sulfur-polymer coating. (a) Solution-based sulfur-polymer deposition on planar silicon for film characterization and electrospun Nano fibrillary films for mercury ion removal (left). b) Images of four sulfur-polymer solutions: DCB, o-xylene, and butanol (water).

9. High Transparence of Sulfur reactive polymer

Scientists have developed a revolutionary technique that improves the high transparence of refractive polymer films via the use of a one-step vapor removal process. These polymers have shown good environmental stability and chemical resistance, which is very advantageous for their use in optical device usage for a long period of time. A high refractive index polymer is produced by polymerizing the evaporated sulfur with several components, which is promptly transported from the elemental sulfur to the polymer. This approach avoids the formation of overly long S-S chains and creates clear non-crystalline polymers that are visible

throughout the visible spectrum (Scheme 7.). Due to the vapor phase process's properties, the high refractive index thin film may be coated not only on silicon wafers or glass substrate materials, but also on a wide range of textured coverings. This thin-film polymer may be the first of its type to achieve an ultra-high refractive index higher than 1.9 [18].



Scheme 7. Sulfur vapor is co-polymerized to form a thin film with a high refractive index.

10. Conclusion:

This study examines the conventional industrial and consumption practices that contribute to the worldwide problem of elemental sulfur utilization and storage. It also aims to shed some insight on the vast range of applications for S-containing polymers. To develop high sulfur polymers, several of the techniques outlined below include using elemental sulfur as a starting point.

- Since elemental sulfur is difficult to dissolve and process, its usage in chemistry is limited. Overcoming these obstacles led to the discovery of sulfur as a new solvent and inverse vulcanization.
- By using the reverse vulcanization process, it is possible to make unique copolymers with a lot of sulfur that can be used in different ways.
- Lithium-sulfur batteries hold four to five times the energy of lithium-ion batteries. This polymer is used to make batteries that are inexpensive, light, and store a lot of energy.
- Reverse vulcanization produces a sulfur polymer with superior process ability and mechanical properties.

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She has been a professor at Eskisehir Osmangazi University, Applied Science, Department Of Chemistry, since 2015. She was born in Turkey. She developed an interest in science and chemistry since childhood. She started her academic education in Chemistry at Anadolu University in the year 1888. She continued the study of Chemistry and Applied Science and in 1992, she wrote a dissertation through which she earned her doctorate in Chemistry and Science. After receiving her doctorate, she decided to work as an Academician at the University of Eskisehir Osmangazi where she performed her duties as lecturer and Researcher. She also published many valuable essays in reputed international journals both in Turkey and abroad that can be an enormous path for other researchers. She had understood the concepts and requisites of academic society. Therefore, she was able to draw theories regarding thiazole and isoxazole. In order to improve the science values, the contribution and publications of her are remarkable and cannot be neglected.

Associate Prof. Lutfullah Ahmadi received the B.s. degrees in Chemical Technology from Kabul Polytechnic University in 1982. Presently he is working as Associate Professor in the Department of inorganic substances industrial Engineering, Faculty of Chemical Industrial Engineering, Kabul Polytechnic University, Kabul Afghanistan.

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Robina Maihan done her bachelor's in chemical engineering in 2013 and start working as an assistant professor in 2014 right now she is doing her master's degree in polymer chemistry at Osmangazi University in Eskisehir, Turkey.