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# Challenges in the Commercialization of Lithium (Li)–Sulphur (S) Batteries †

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***Abstract***

*Li-S batteries can be used in drones owing to their high gravimetric energy density (2600 Wh/kg of sulphur) as well as the high theoretical specific capacity values (1674 mAh/g) values. Most of the ground breaking inventions in energy sources occurred during war time (1$^{st}$ September 1939-2$^{nd}$ September 1945), including the discovery of the famous nuclear fission process that lead to the origin of the little boy and the fat man. Such innovation is expected in battery technology surpassing that of the Israeli iron dome and the design of energy efficient and high precision drones. The use of battery technology in drones for application in war is upcoming as seen in Ukraine and Gaza by the invading disruptive forces. The current review article is aimed at developing the advanced and state of the art cathode materials based on S modified carbon materials leading to sustainability, safety and cost effectiveness. As we are currently in the war time, this research on the development of cathodes for Li-S batteries is highly relevant for defence as well as for mobile and transportation application in the war zones.*

**Keywords:** Cathodes, Carbon-sulphur composites, Kinetics, Shuttle effect, Polysulfide intermediates, Gravimetric energy density, Specific capacity, Rate capability, Cycling stability safety

## INTRODUCTION

The problem human societies are facing owing to scarcity of clean energy sources needs no introduction. Extensive dependence on fossil resources for energy needs, in all most all human endeavours, has already caused an irreversible damage to the environment we live. So to remedy the mistakes of the past, there is imperative that we develop clean energy sources that will have the least carbon foot print and that we prevent any further increase in the global average temperatures. As a solution to the twin problems of scarcity of clean energy sources and environmental damage, it is an urgent need to move towards the use of electrochemical energy storage systems, namely, batteries, for stationary as well as mobile applications. Among various types of batteries, the lithium-sulphur batteries stand out owing to their high theoretical specific capacity (1674 mAh/g) as well as high gravimetric energy density (2600 Wh/kg of sulphur) values [1–5]. This has been the subject of the current study. Although, the theoretical specific capacity as well as energy density values derived from the Li-S batteries are very high, these theoretical values could hardly be achieved due to many kinetic constraints involved in the redox chemistry of sulphur and also due to the famous shuttle effect caused by the lithium polysulfide intermediates. Intense research and development efforts are going on, in the field of batteries in general, and in the area of Li-S batteries in particular. A web of science search with the keywords namely, Lithium-sulphur batteries yields 15,063 results in total. Out of them, 460 results are only from the year 2024 alone, ie in

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the past four months alone (1/1/2024 to 9/5/2024). This shows the intensity of research going on in the arena of Li-S batteries and especially, in developing appropriate cathode materials, which is indeed challenging. A growing trend in the research of Li-S batteries as a function of time, especially, during the past decade can be seen from the data of web of science converted into knowledge as shown in Figure 1.

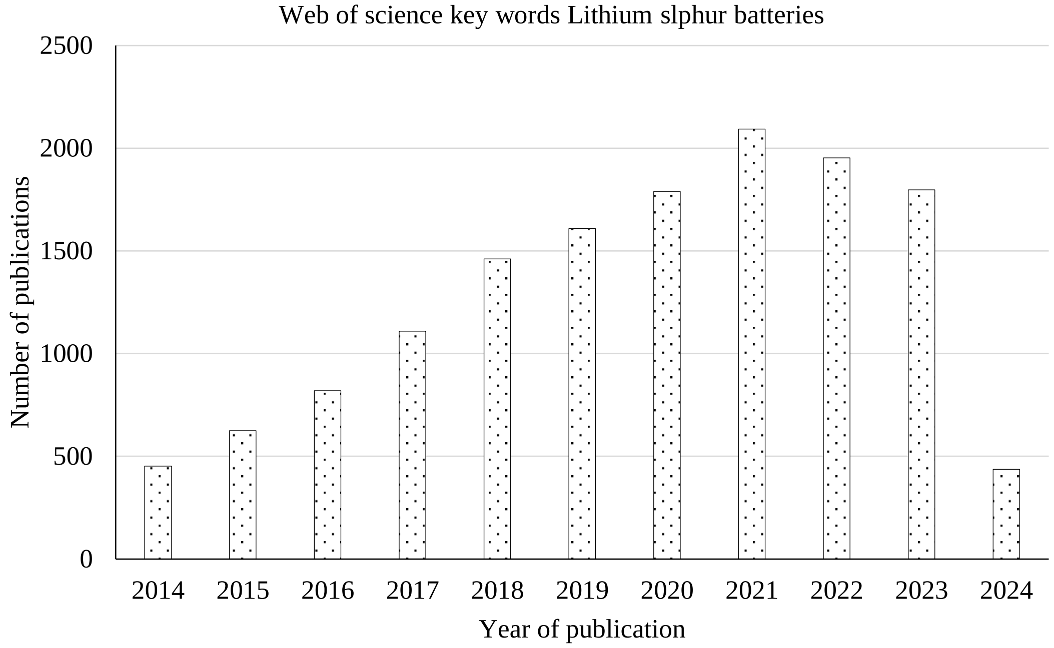



**Figure 1.** Intensity of research in the field of lithium sulphur batteries signifying the important of the research area (from the Web of Science).

Globally, there is a renewed interest in the use of electrochemical energy storage and conversion devices owing to their environmental friendliness and sustainability. Among various batteries, the lithium – sulfur (Li-S) batteries stand out. Obviously, this is because of the cost competitiveness as well as the sustainability due to the low cost, abundance and non-toxicity of sulfur. Move over, the electrochemical performance of the Li-S batteries is 3-5 fold higher than the celebrated commercial Li ion batteries (LIBs). However, there are major obstacles, that need to be surmounted before reaching the goal of the commercial scale utilization of Li-S batteries and the current research article is based on such goal. As high as 1674 mAh/g of theoretical capacity can be achieved, in principle, from the Li-S batteries. Moreover, among all the known rechargeable Li-batteries, the highest theoretical energy density (2600 Wh/kg) is derived from Li-S batteries. This makes the Li-S batteries the obvious choice as future sustainable energy conversion devices. However, owing to serious kinetic and diffusion constraints, the theoretical values of capacity and energy density have not yet been realized. One of the aims of the researchers in the field is to overcome such kinetic and diffusion barriers and to attain the capacity and energy density values near to the theoretical values by developing Sulfur modified carbon nanomaterials as cathode materials for Li-S batteries. Use of carbon material as support facilitates high loading of S per geometric cathode area. Moreover, carbon materials facilitate electronic conductivity between the carbon surface and the sulfur species contained in the porous architecture of carbon materials. Unconventional carbon materials, as those produced from biomass, lignin and microporous activated carbon materials modified with sulfur doped carbon dots will form promising cathode materials for the Li-S batteries to realize the electrochemical performance superior to the advanced LIBs [31].

Clearly, energy security and independence is one of the foremost priorities of any nation and it is more so with the nation India. With the growing uncertainty and instability in the accessibility to fossil based resources and the environmental deterioration and climate change associated with their indiscriminate use, it is imperative that sustainable and clean alternate energy sources are developed. Li batteries are one such alternate energy sources to petroleum based resources for both domestic and transportation applications. Among, various Li batteries, the Li-S batteries standout owing to the very high theoretical capacity (1672 mAhg-1) as well as energy density (2600 wh $kg^{-1}$). However, such high values of theoretical capacity and energy density which are nearly 3-5 fold could have never been realized owing to the kinetic and diffusion constraints involved in S cathodes of the Li-S batteries. Though the fundamental research studies on Li-S batteries were started nearly four decades ago, the obstacles faced, namely, the diffusion and kinetic constraints reduced the derivable capacity from 1674 to 1000 $mAhg^{-1}$ [4]. However, there have been a renaissance and renewed interest in the R and D efforts of Li-S batteries owing to the advances in materials science and nanotechnology leading to surmounting the obstacles and making the utilization of sulfur highly efficient at the cathode. There is a huge hope for the commercialization of Li-S batteries for the application in electric vehicles (EVs) as well as for electronic devices (lap tops and mobile phones). Use of carbon nanomaterials as host to S by impregnation, doping or anchoring or deposition has been a very promising strategy for the effective utilization of S as the cathode materials for Li-S batteries. There has been advances in the synthesis of high surface area microporous activated carbon materials that could be functionalized with sulfur that serves as potential and sustainable cathode material. The present research effort is towards attaining the sustainability in the cathode materials for Li-S batteries, by designing effective synthetic methodologies for the S modified carbon materials resulting in the realization of the attainment of capacity as well as energy density values close to the theoretical values. Moreover, advanced synthetic techniques like sonochemical irradiation, and Reaction under Autogenic Pressure at Elevated Temperature (RAPET) method will be useful for producing S modified high surface area activated carbon materials. The porous architecture of the carbon materials allows the penetration of the electrolyte and also prevents the loss of the reduction products of sulfur. The distribution of sulfur is dependent on the porous structure of the carbon host material and its uniform distribution facilitates uniform response of the electrode. Use of such unconventional carbon supports for sulfur (either elemental S or S doped carbon dots) facilitate the high loading of S per geometrical cathode area.

The objective of composing the review article on the development of cathode materials for Li-S batteries is to provide an insight on the application of nano sulphur functionalized porous carbon materials from unconventional feedstock like the lignocellulosic biomass produced using exotic synthetic techniques like sonochemistry, microwave irradiation, plasma irradiation and RAPET. The sulphur containing carbon materials with unique structural and textural properties and nanoarchitecture form potential cathodes in Li-S batteries. Such carbon – sulphur composites are hypothesized to overcome the typical problems, including the poor kinetics of the S redox process and the famous challenge of the shuttle effect, leading to the realization of attaining the theoretical values of the specific capacitance (1674 mAh/g) as well as gravimetric energy density (2600 Wh/kg of sulphur). Based on the state of the art literature, such materials were recommended for designing the commercially adaptable Li-S batteries. Use of carbon materials, and sulphur which are abundant in nature and are of low cost makes the Li-S batteries economical as well as environmentally friendly. A brief history of the challenges in the Li-S batteries, like the poor redox kinetics at the cathode as well as the well-known hurdle of the incomplete reduction of the lithium polysulfide intermediates formed at the cathode and their migration (shuttle effect) is provided along with possible solutions to surmount the same. As there will be no $CO_2$ emissions in the electrochemical reactions taking place at the Li metal anodes and sulphur-carbon composite cathodes, these Li-S batteries resulting in high gravimetric energy density and specific capacity have many fold applications in the realms of energy and environment. In addition, there is an urgent need to move from the use of highly polluting fossil based fuels namely petrol and diesel to the non-polluting and circular economy energy sources like electric vehicles that use batteries as the energy storage medium.

## CHALLENGES IN THE PERFORMANCE OF CATHODES OF Li-S BATTERIES

Intense scrutiny is occurring on the cathode materials for the lithium (Li) – sulphur (S) batteries in the R and D establishments [1–32]. This is due to the high gravimetric energy density (2600 Wh/kg of sulphur) as well as the high specific capacity (1674 mAh/g of sulphur) values that could be delivered by the Li-S systems. Moreover sulphur being available and cheap it leads to sustainability. In addition, the use of sulphur can be drastically minimized by addressing the problem of the electrical insulting nature of the sulphur by depositing this exotic material on the magic material, namely, the carbon materials. Carbon materials are unique. They are abundantly available, with tunable, surface functionality, surface composition, texture, structure and morphology. Carbon materials being electrically conductive forms the ideal host material to sulphur and a beautiful combination of Carbon and sulphur leads to a promising cathode materials for Li-S batteries. The chances of overcoming the kinetic constrains as well as the famous problem of the shuttle effect are very high in the use of carbon – sulphur composites as cathode materials. Professor Doron Arbach’s group published a land mark paper on the use of $LiNO_3$ as an additive in overcoming the problem of shuttle effect [5]. In the same paper, Professor Aurbach’s group developed state of the art method to fabricate the air sensitive samples of Li for studying the surface chemistry using FT-IR and XPS. Much of the details of the surface chemistry on either the anode or the cathode are missing. Such studies will provide solutions in surmounting the kinetic constraints in realizing the theoretical values of the gravimetric energy density as well as the specific capacity values and the current review article precisely aims at developing such unconventional state of the art methodologies for the development of cathode materials and their systematic characterization.

The plateaus in the discharge curve of the Li-S batteries with the formation of lithium polysulfide intermediates is schematically represented in Figure 2. The high theoretical specific capacity value of 1674 $mAhg^{-1}$ can only be realized when all the polysulfides are successfully converted to $Li_2S$. However, in reality, the solubility of lithium polysulfides in the electrolyte makes them move away from the cathode before undergoing complete reduction. This leads to severe mass loss and the accompanied low values of specific capacity as well as the gravimetric density that are realizable. This problem is well known in chemical literature as the shuttle effect. For better insight, the phenomena of shuttle effect is schematically depicted wherein the reaction intermediates, lithium polysulfides drift away via diffusion into the anode leading to severe mass loss as well as poor cell performance [7].

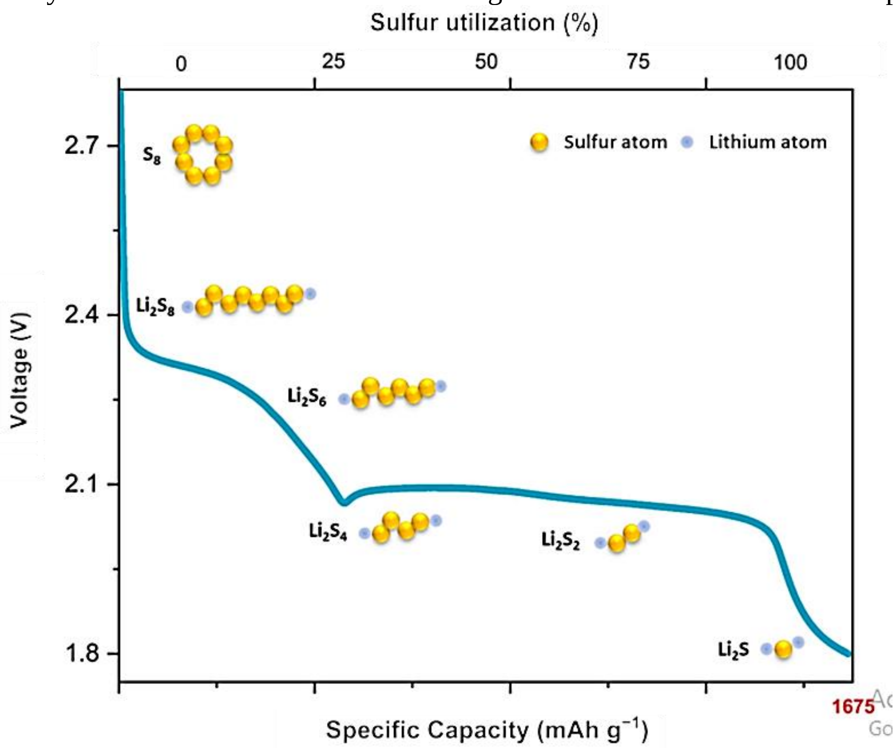



**Figure 2.** Typical discharge curve of Li-S batteries [Adapted with permission from reference 20,

MDPI].

Typical problems that hinder the commercialization of the Li-S batteries include, the poor kinetics of the redox processes at the cathode, insulating nature of the sulphur, the volume expansion of sulphur leading to safety concerns and the well know issue with the migration of the lithium polysulfides from the cathode prior to complete reduction (the shuttle effect). Provided these issues are addressed, the specific capacity values as well as the energy density values close to the theoretical values can be attained with good cycling stability and outstanding electrochemical performance leading to commercial scale exploitation of these electrochemical energy storage devices. This would address several challenges facing human societies currently, namely, lack of petroleum alternatives, environmental pollution to autoexhaust emissions, safety and security concerns in the field of defence and many more.

## UNCONVETIONAL CARBON-SULPHUR COMPOSITES AS CATHODE MATERIALS FOR Li S BATTERIES

Among various host matrices for S, namely, MXenes [17], linear sulphur copolymers [18], metal organic frame works [22], the carbon materials standout owing to the ease of synthesis and peculiar adsorption properties for the reaction intermediates, namely, the lithium polysulfides, formed at the cathode. [4]. In particular, the recent trend in the exploitation of Carbon-sulphur composites as cathode materials for Li-S batteries, is the use of porous and functionalized carbon materials from unconventional sources like the biomass. Outstanding performance of the Li-S batteries with the use of carbon materials produced from biomass and subsequently loaded with sulphur has been reported widely in literature [25–29]. The data shown in Table 1, indicate the potential of the activated carbon materials derived from biomass as host materials for S for the fabrication cathodes for Li – S batteries.

**Table 1:** Biomass derived porous carbon materials as sustainable matrices for sulphur in the fabrication of Li-S batteries [7]

| **Biomass source for activated carbon material** | **Initial discharge capacity ($mAhg^{-1}$) with rate** | **Reference** |
|---|---|---|
| Garlic peel | 1087 at 0.1 C | 25 |
| Jamun seed | 1330 at 0.1 C | 26 |
| Durian shell | 723 at 0.5 C | 27 |
| Pomato peel | 1534 at 2 C | 28 |
| Sugar cane bagasse | 815 at 1 C | 29 |

Biomass forms an abundant source of lingo-cellulose that could be carbonized to highly porous activated carbon materials. Forming composites of such biomass derived carbon materials with elemental S is an innovative strategy towards attaining the theoretical values of the gravimetric energy density and specific capacitance.

Field emission–scanning electron microscopy (FE-SEM) images of activated carbon derived from orange peels and loaded with sulphur showed homogeneous distribution of as high as 52.13 wt.% on the porous network on carbon material [7]. This shows the potential of special carbon materials prepared using chemical activating agent (KOH) resulting in porous carbon materials that forms a promising host to sulphur resulting in advanced cathode materials [7]. However, for the production of carbon materials unconventional chemical activating agents like organic acids and the alkali metal salts there of need to be exploited for the cleaner synthetic methodologies as substitute to the corrosive compounds like KOH, NaOH and $K_2CO_3$ [33]. Not only the synthetic strategies, but the analytical tools for the characterization of the carbon–sulphur composites were also evolving. Field emission–transmission electron microscopy (FE-TEM) with elemental mapping using electron energy loss spectroscopy (EELS) facilitated viewing of the elements carbon and sulphur distributed homogenously on the surface in the activated carbon – sulphur composite produced using orange peel waste as the benign, cheap and sustainable feedstock.

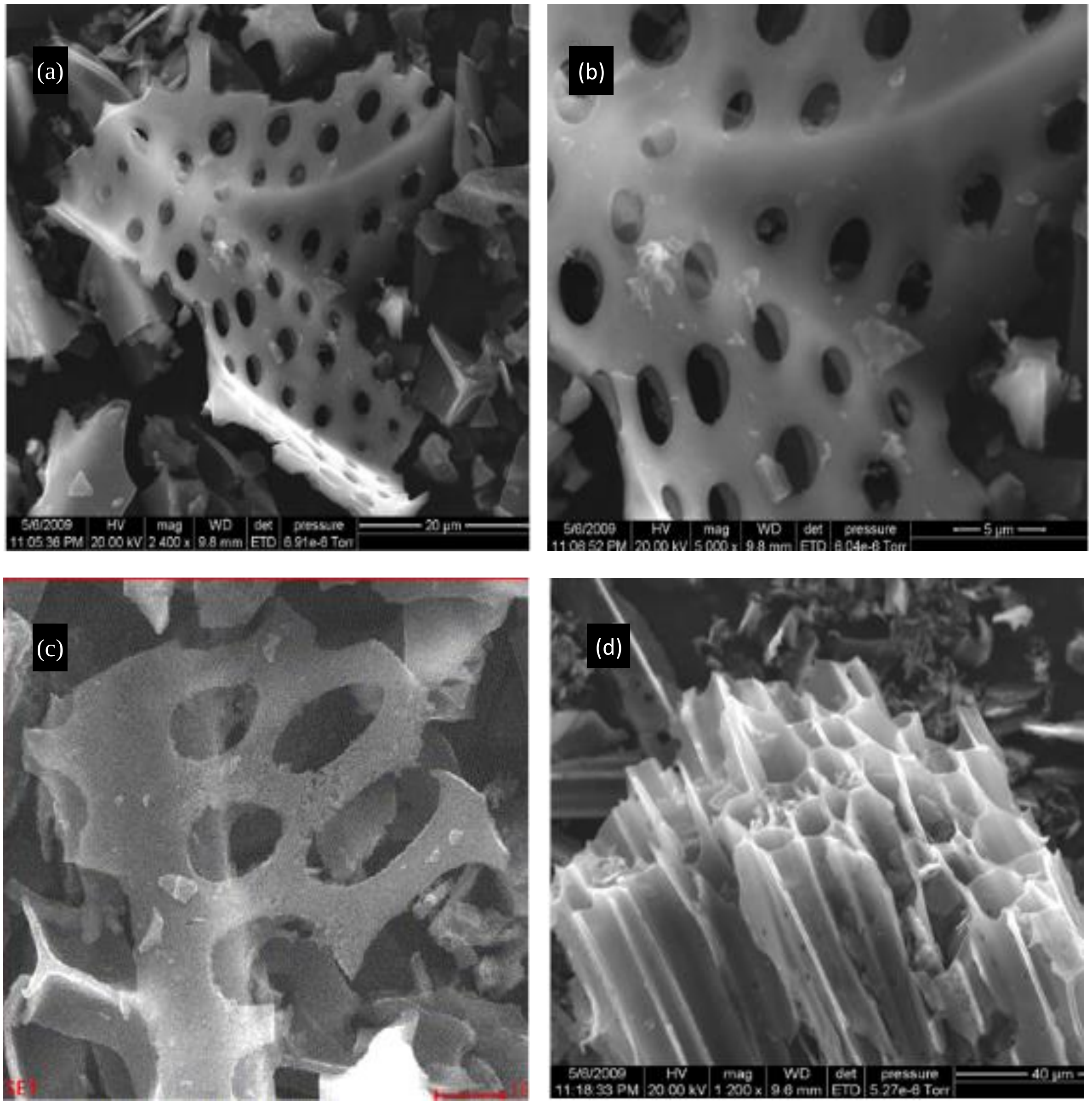
 

Such homogeneous distribution of sulphur on conducting carbon solves the problem of the insulating nature of the sulphur resulting in exceptionally high values of specific capacitance as well as unusual rate capability and cycling stability [7]. Intense scrutiny of the state of the art literature, clearly signal the promise of Li-S batteries for future use in drones in war zones as well as for mobile and domestic and commercial applications owing to the high values of energy density, specific capacitance realizable provided the typical problems in the operation of cathode materials are surmounted. The current review exactly aims at providing the insight needed for surmounting the typical problems in the cathode materials, by developing sustainable carbon – sulphur composites. Undoubtedly, sonochemicals strategies developed by Professor Aharon Gedanken's group facilitate nanosized sulphur particles adhere strongly to the carbon surface increasing their activity many a fold [34]. Professor Aharon Gedaneken's group developed state of the art microwave and RAPET methods too for producing carbon materials that could be useful for application in forming composites with S [35, 36].

## FUTURE PERSPECTIVES:

Layered carbon graphitic structures (graphite recovered from used Li ion batteries for instance) served as promising host for S and the resulting cell performance from such cathodes is remarkable [15]. Based on the encouraging results from literuature, it is hypothesised, that the layered carbon materials from sustainable biomass like the stems of *Calotropis gigantea* and the spikes of the *Borassus flabellifera* (Figure 3) forms a potential host for sulphur for application as cathode materials for Li-S batteries. The SEM images of the activated microporous carbon materials with a peculiar layered structure derived from the stems of *Calotropis gigantea* and the spikes of the *Borassus flabellifera* were shown in (Figure 4). Such exotic layered and porous architectures serves as a promising host material for the homogeneous distribution of nanoforms of S enhancing the S utilization and there by facilitating the realization of achieving the values of energy density and specific capacity values nearing the theoretical values. Such layered prorous structured high surface area active carbon material will act as potential adsorbents for polysulfide intermediates formed during the redox reactions at the cathode there by arresting the shuttling effect [13].


**Figure 3.** Stems of *Calotropis gigantea* (a) and the spikes of the *Borassus flabellifera* (b) [30–32].


(a)
(b)
(c)
(d)
20 µm
5 µm
40 µm

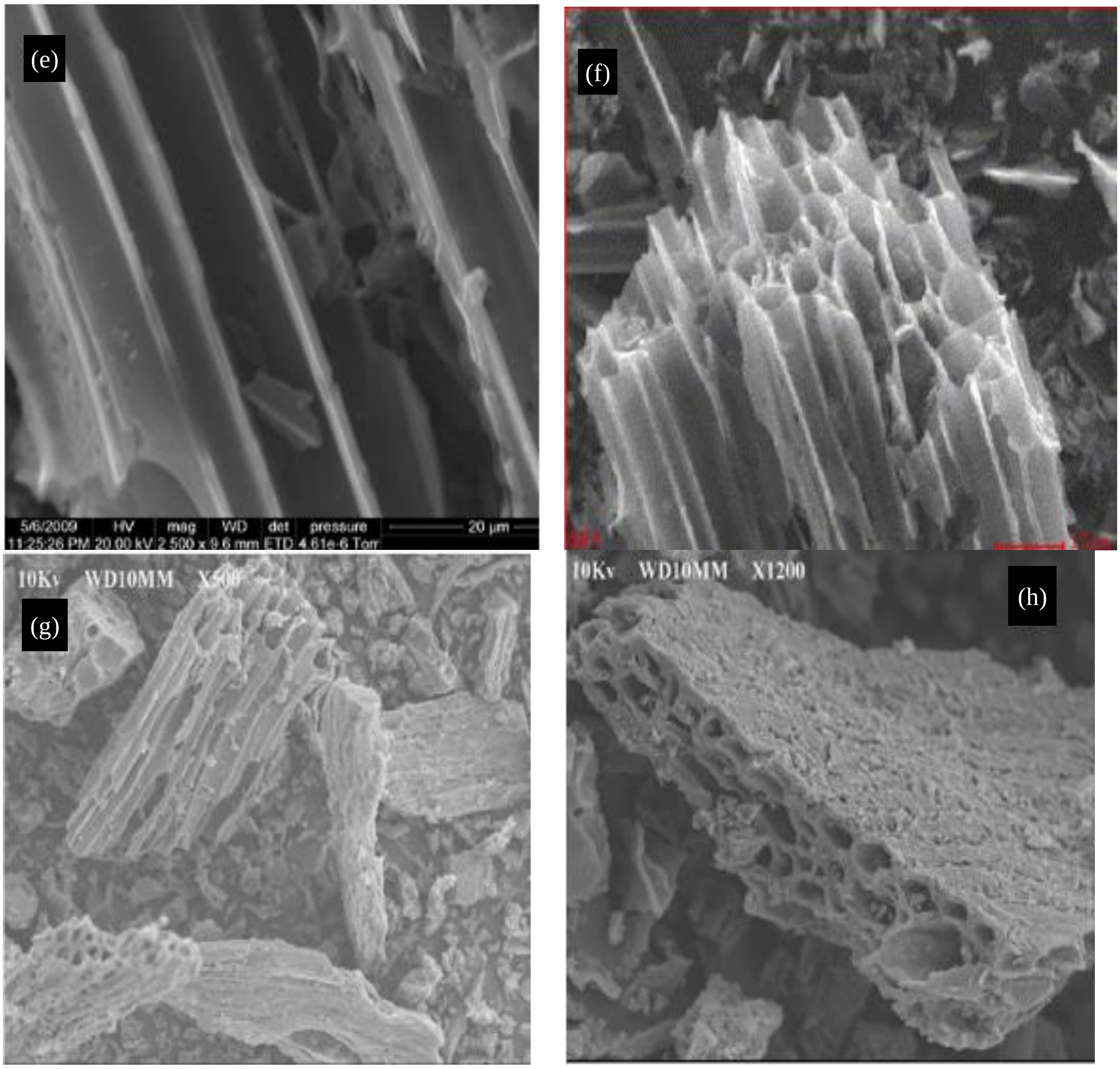



**Figure 4.** SEM images of the activated microporous carbon materials with a peculiar layered structure derived from the stems of *Calotropis gigantea* (a–f) and the spikes of the *Borassus flabellifera* (g) and (h) [30–32].

The subject of development of carbon sulphur composites as cathode materials for Li-S batteries have aroused enthusiasm world-wide owing to the potential of these materials to over-come the classical problems of poor kinetics at the cathode as well as the well-known shuttle effect. The innovation is that the use of porous activated carbon materials with high surface area from biomass leads to sustainability. Such a study has resulted in several patents [30] and the current insight in that direction is hypothesized to result in a breakthrough. The carbon materials proposed to be used as host materials for sulphur have the potential of adsorbing the lithium polysulfides that would result in the complete reduction of the polysulfide intermediate species leading to the realization of the theoretical limits of high gravimetric density (2600 Wh/kg of S) as well as the high specific capacity (1674 $mAhg^{-1}$) as shown in Figure 5. These aspects are new in literature.

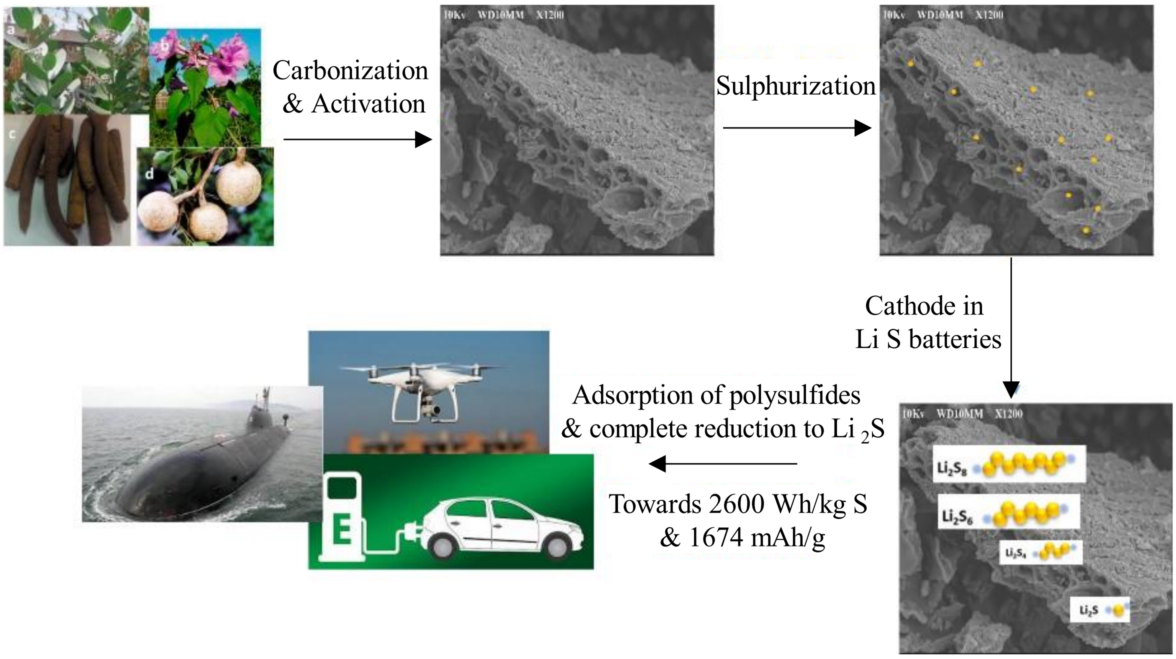



**Figure 5.** Schematic representation of the innovative strategy to attain the theoretical limits of gravimetric density and specific capacity of the Li-S batteries.

## CONCLUSION

The problem of researching into Li-S batteries needs no emphasis on its significance. Human societies are currently facing the challenge of constraints and shortages of energy sources, especially, for mobile applications in transportation sector. Li-S batteries are superior to the commercial Li ion batteries by a factor of five in terms of electrochemical performance (gravimetric energy density 2600 Wh/kg of S as well as specific capacity 1674 mAh/g). However, owing to the poor kinetics of the redox process at the cathode, as well as due to the incomplete reduction of sulphur due to the formation and migration of the lithium polysulfides, the practical values of the cell performance are much lower than expected. The current review article precisely addresses these issues by proposing better materials that are cheap and sustainable. Use of such unconventional cathode materials yield a breakthrough in Li-S batteries resulting in the development of human societies by providing alternatives to petroleum. As a result, the twin problems of green energy needs and clean environment could be solved. Moreover, as we are in war time, for national defence and security too battery technology, especially, the lithium sulphur technology is vital.

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**†Dedicated to the legendary Jewish chemist Professor Aharon Gedanken on the occasion of his 84th birthday on 24th December 2024.**

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