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ELECTRICALLY CONDUCTIVE FILMS - CHARACTERISATION OF WEIGHT-REDUCED POLYMER FILMS FOR USE IN A BIPOLAR LITHIUM-SULPHUR BATTERY

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ABSTRACT

With a theoretical energy density of around 2600 Wh kg-1 and a specific capacity of approx. 1675 mAh g-1, the lithium-sulphur battery (Li-S battery) is a promising technology for future energy storage solutions. Lithium-sulphur batteries currently achieve specific energy densities of between 400 and 600 Wh kg-1 in practical applications, making them an attractive alternative to lithium-ion batteries. Despite their advantages, technical challenges such as the dissolution of polysulphides in the electrolyte and the low cell voltage are significant obstacles to the commercialisation of this technology. In the "BiPoLiS" project (development of lithium-sulphur bipolar batteries), these challenges are addressed by combining lithium-sulphur battery technology with a previously undescribed bipolar structure. The focus of this article is on the development of weight-reduced polymer conductor foils, which should contribute to an improved battery packing density due to the planar layer structure - compared to single cells.

Experimental investigations were carried out to develop an electrically conductive, flexible and weight-reduced film suitable for use in lithium-sulphur batteries. The investigations were based on compounds of styrene-ethylene-butylene-styrene (SEBS) and polypropylene (PP), whereby various mixing ratios were tested. The optimum mixture was found with a ratio of a SEBS compound to a PP compound of 30:70, which had an electrical in-plane conductivity of around 346 S m-1 and a through-plane conductivity of around 60 S m (-1). The flexible film was produced using a calendering process, whereby a thickness of approx. 0.2 mm was achieved.

Thanks to the innovative combination of bipolar design and polysulphide-free sulphur cathode, the lithium-sulphur battery can offer significant advantages in terms of energy density and manufacturing costs. This technology could play a decisive role, particularly in the field of electromobility and industrial energy storage. Future work should focus on optimising the process parameters for reducing the film thickness and investigating the long-term stability in order to exploit the full potential of these new materials and enable their commercial applications.

Keywords: Bipolar Lithium-Sulphur Battery, Energy Storage Solutions, Polymer Conductor Foils

1. INTRODUCTION

The lithium-sulphur battery is a promising technology for application-orientated energy storage solutions of the future. This is mainly due to its high theoretical energy density of approx. 2600 Wh kg^{-1} and specific capacity of approx. 1675 mAh g^{-1} Zhang (2013), Manthiram et al. (2013). In practical applications, it currently achieves specific energy densities of between 400 and 600 Wh kg^{-1} and is

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therefore significantly higher than the lithium-ion battery (150-250 Wh kg^{-1}) Manthiram et al. (2013). Sulphur is one of the 20 most abundant elements in the earth's crust, making it a cost-effective cathode material that is available worldwide without the need for complex global transport networks Manthiram et al. (2013). In addition to the aforementioned advantages, several technical challenges are responsible for the fact that commercial application with long runtimes is only imminent in the future. These limitations include the dissolution of polysulphides diffusing through the electrolyte, which reduces cell voltage and cycle life Manthiram et al. (2013), Zheng et al. (2011).

As part of the "BiPoLiS" project (development of lithium-sulphur bipolar batteries), these challenges are being met by combining lithium-sulphur battery technology with the design of bipolar batteries. A polysulphide-free sulphur cathode and a weight-reduced conductor foil, which is the focus of this article, are being developed by Whitecell Eisenhuth and the IPF Dresden. By increasing the energy density per volume through the bipolar structure (instead of individual cells), a lithium-sulphur battery prototype is then built, which has not yet been described in the literature and is also characterised by its low weight for mobile applications.

2. STATE OF THE ART

The basic principle of the lithium-sulphur battery is shown in Figure 1 The cell consists of a lithium anode and a sulphur cathode, which are connected to each other via a load resistor or a voltage source. An organic electrolyte also spatially separates the anode and cathode Manthiram et al. (2013).

Figure 1

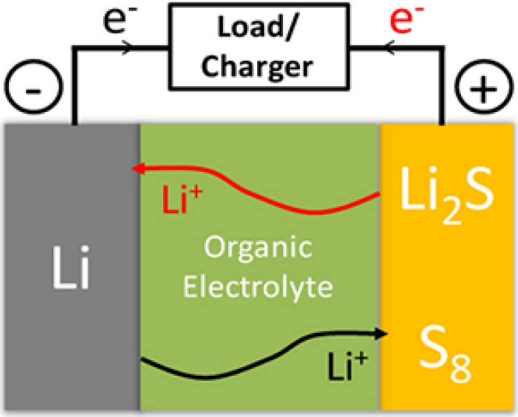

Figure 1 Schematic Diagram of the lithium-Sulphur Battery (LiS) Manthiram et al. (2013).

During the discharge process, elemental lithium is oxidised at the lithium anode according to *Eq. 1* Nazar et al. (2014).

$$\mathrm{Li} \rightarrow \mathrm{Li}^+ + \mathrm{e}^- \qquad \text{(Eq. 1)}$$

On the cathode side, sulphur is reduced in chained intermediate steps, resulting in the formation of partially soluble polysulphides (*Eq. 2*) Nazar et al. (2014).

$$\mathrm{S}_8 + 16\,\mathrm{e}^- + 16\,\mathrm{Li}^+ \rightarrow 8\,\mathrm{Li}_2\mathrm{S} \qquad \text{(Eq. 2)}$$

The overall reaction is described below (*Eq. 3*) Nazar et al. (2014).

$$16\ \mathrm{Li} + \mathrm{S}_8 \rightarrow 8\ \mathrm{Li}_2\mathrm{S} \qquad \text{(Eq. 3)}$$

The potential of the lithium-sulphur battery lies in its three to five times higher energy density compared to lithium-ion batteries. This makes them particularly attractive for electromobility and industrial energy storage. The high theoretical capacity is based on the oxidising capacity of sulphur, which can absorb up to two electrons per atom. Despite this, considerable technical challenges currently represent a barrier to the commercial market maturity and acceptance of the lithium-sulphur battery Zhang (2013), Manthiram et al. (2013).

One of the main obstacles is that lithium-sulphur batteries only have a low cell voltage of around 2.1 V compared to 3.7 V for lithium-ion batteries. This results in an almost doubling of the cells and interconnections for the same electrical battery voltage, which increases the weight and complexity of the battery pack Nazar et al. (2014).

The second major obstacle is the solubility of lithium polysulphides (Li_2S_8, Li_2S_6, Li_2S_4, Li_2S_2), which are formed as intermediate species in the cathode reaction (*eq. 2*). They influence the charging and discharging performance. Figure 2 shows a qualitative curve of the electrical voltage of a lithium-sulphur battery over the capacity with significant voltage drops as a result of the solubility of various polysulphides Manthiram et al. (2014).

Figure 2

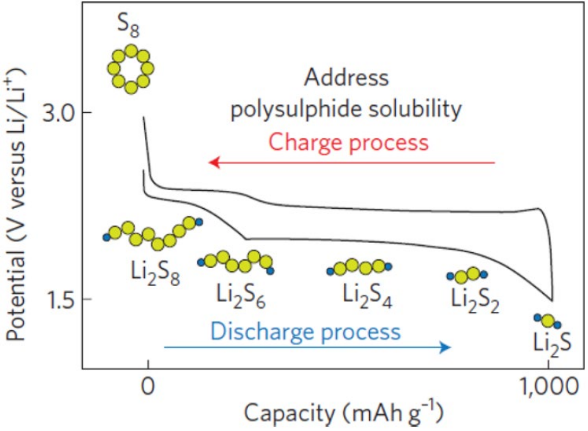

Figure 2 Electrical voltage curve during charging and discharging of the LiS battery Manthiram et al. (2014)

Polysulphides are soluble in most electrolytes and can diffuse through them to the lithium anode, where they are no longer part of the active cathode mass. At the lithium anode, they trigger undesirable side reactions, such as reduction to elemental sulphur. This so-called "shuttle effect" favours self-discharge and reduces battery efficiency. Zheng et al. (2011), Abdollahifar et al. (2022)

3. TECHNOLOGICAL INNOVATIONS AND SOLUTIONS IN THE "BIPOLIS" PROJECT

The "BiPoLiS" project overcomes the aforementioned challenges of lithium-sulphur battery technology through the structural bipolar design of the battery cells with polysulphide-free sulphur cathodes.

One of the work packages within the project is the development of the sulphur cathode, which prevents the dissolution of polysulphides in order to improve the cycle stability and service life of the battery.

In contrast to the established battery concepts, in which the individual cells are installed in individual housings, the bipolar plate to be developed enables the direct connection of individual cells without the need for complex connection technology for the electrical circuitry.

These electrically conductive films, which are coated on the one hand with cathode active material newly developed by project partners and on the other with anode active material, form a battery stack when stacked with separator films between them. With the aid of sealing adhesive, the bipolar plates are bonded at their outer edges so that neither active material nor liquid electrolyte can escape into the environment.

Thanks to the bipolar design and the resulting reduction in system weight, the new combination is intended to unite the advantages of both technologies and enable a higher energy density at lower manufacturing costs due to shorter process chains.

For applications in the mobility sector, bipolar stack construction can be of decisive importance due to the weight and cost savings.

The experimental work in this article focusses on the material research of bipolar conductive films based on polymers and electrically conductive graphitic fillers as well as on the identification of a promising manufacturing process.

4. EXPERIMENTAL INVESTIGATIONS

As part of the project, Whitecell Eisenhuth has carried out a study to develop a conductive, flexible and weight-saving electrically conductive film with a low filler content that is suitable for use in a lithium-sulphur battery. The thickness of the film should be 0.1-0.3 mm, with a minimum electrical conductivity through the film plane of 1 S m^{-1}. The low thickness of the film should make it pliable and flexible.

The procedure was carried out in two stages:

1) Development of a promising compound
2) Testing of the most suitable compound candidate in the calendering process

SEBS is a thermoplastic polymer characterised by elasticity similar to rubber materials and flexibility over a wide temperature range (-50-100 °C). It is also stable against many acids, bases and alcohols. The SEBS used in this work was directly mixed with 30 wt% conductive carbon black in its original state Drobny (2007).

As a widely used thermoplastic polymer, polypropylene is a popular and cost-effective binder for fillers. It is easy to process in extrusion and injection moulding. It also has good heat resistance up to around 100 °C and is resistant to acids, bases and organic solvents [8]. In this test series, "EEE" (a standard injection moulding formulation from Whitecell Eisenhuth) was used, which uses polypropylene as a binder and contains 86 wt% graphitic filler.

Both polymers used (polypropylene and SEBS) are miscible and this combination is frequently used in industry due to its versatile properties Drobny (2007). To improve the processability of the test series, a small amount of silicone lubricant (GM) was added to the compound. During the manufacturing process (compounding, injection moulding, calendering), this lubricant is intended to form a separating layer between the movable metal of the process technology and the compound to be processed.

In the first stage, a test series was set up in Table 1 based on styrene-ethylene-butylene-styrene (SEBS) and polypropylene. The SEBS compound and the EEE compound (PP) were tested in their pure form without mixing and without the addition of silicone lubricant (GM) (materials 1 and 8). Subsequently, 50-80 wt% EEE compound was gradually added to the SEBS compound (material 2-7).

Table 1

Table 1 Test Series for Compound Characterisation

Material no.	Designation	SEBS compound / %	EEE (PP compound) / %	Silicone GM / g/kg$_{Compound}$
1	SEBS/EEE+GM 100/0+0	100	0	0
2	SEBS/EEE+GM 100/0+20	100	0	20
3	SEBS/EEE+GM 50/50+20	50	50	20
4	SEBS/EEE+GM 40/60+20	40	60	20
5	SEBS/EEE+GM 30/70+20	30	70	20
6	SEBS/EEE+GM 20/80+20	20	80	20
7	SEBS/EEE+GM 0/100+20	0	100	20
8	SEBS/EEE+GM 0/100+0	0	100	0

Two types of test specimens were then produced from the materials in an injection moulding machine. On the one hand, flow spirals Figure 3, the length of which is used to infer the viscosity achieved as a function of the processing temperature and, on the other hand, rectangular test plates Figure 3. These were used to assess the electrical conductivity.

Figure 3

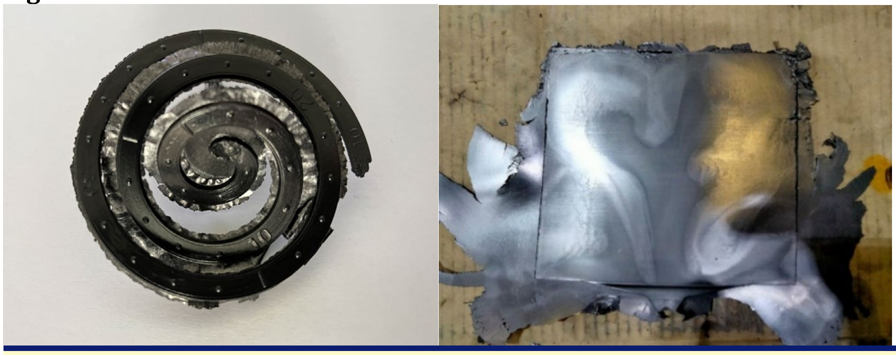

Figure 3 Viscosity Flow Spiral (Left), Conductivity Test Specimen (Right)

The electrical in-plane conductivity was determined using a 4-point contact electrode according to Uhlir Uhlir (1955). The results of the inplane conductivity are shown in Figure 4 On the one hand, the SEBS was found to be less conductive (1.47

S cm^{-1}) than pure EEE (approx. 3.85 S cm^{-1}). On the other hand, there was hardly any change in conductivity up to an admixture of 50 wt% EEE in the SEBS compound.

Figure 4

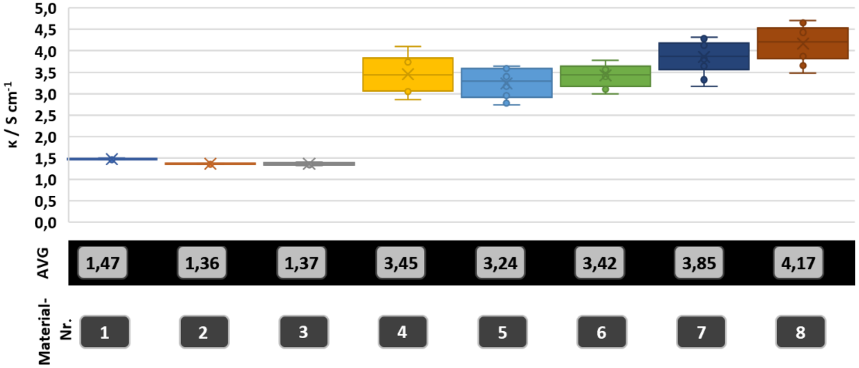

Figure 4 Results of the Specific Electrical Inplane Conductivity Κ

From an admixture of 60 wt% EEE compound, the electrical conductivity κ increased sharply with higher intrinsic dispersion and almost reached the conductivity of the EEE with GM additive (20 g kg^{-1}). From this it can be concluded that the EEE component forms a continuous phase from 60 wt% and the SEBS component is dispersed in it.

The electrical through-plane conductivity is subsequently determined using the Kelvin method. This method determines the specific resistance of the compounds by measuring the resistance across the film thickness Skotheim & Reynolds (Eds.). (2007).

For the investigation, the sample plates were measured once unprocessed and once the surface was decoated by blasting with glass beads in order to remove polymer layers near the surface, which can form on the sample during cooling and mainly influence the throughplane conductivity.

The results are shown in Figure 5 The resistance of the untreated samples gradually increases the more EEE compound is added to the SEBS. The opposite is true for the uncoated samples. The resistance decreases as the proportion of polypropylene compound increases.

The evaluation of the flow spirals is also implemented in the diagram. At a proportion of 70 wt% EEE in the SEBS compound, the flow spiral reaches a maximum length and thus the lowest viscosity within the test series.

Figure 5

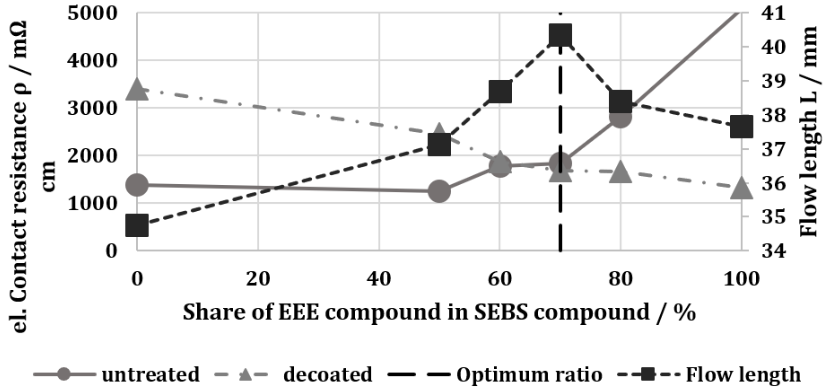

Figure 5 Electrical Resistance Through the Foil Plane and Visualisation of the Flow Lengths

As the resistances of both the untreated and the decoated test specimens are at a low level in this mixing range and cross, the mixing ratio SEBS:EEE: 30:70 is selected as the optimum point in the test series. The through-plane conductivity at this point is around 60 S m^{-1} and well above the minimum requirement of 1 S m^{-1}. The total filling level of conductive filler is approx. 50 wt% at the optimum point.

In the second stage, the most suitable compound candidate (SEBS/EEE+GM 30/70+20) was tested in the calendering process Figure 6.

Figure 6

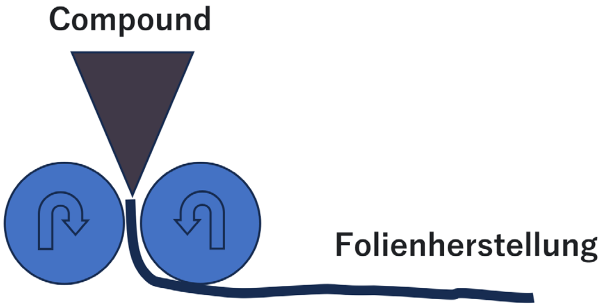

Figure 6 Principle Illustration of the Calendering Process

The compound is fed from above onto two counter-rotating conveyor rollers, which are heated to 220 °C and melt the compound between them. The film produced is then discharged through a set gap distance of 0.2 mm at a speed of 50 cm min^{-1} and stored on rolls.

In the next step, the film is cut to a test geometry Figure 7 using an Epilog Zing 24 laser cutter.

Figure 7

Figure 7 Conductor Film from Calendering Process (Left), Film Thickness Measurement (Right)

The first tests with the compound at the optimum (SEBS/EEE+GM 30/70+20) show a thickness of 0.21 mm. In subsequent tests, attempts are being made to reduce the film thickness to 0.1 mm by modifying the parameters (process temperatures, conveying speed).

5. CONCLUSION AND FUTURE PROSPECTS

As part of this project, an investigation was carried out to develop a conductive, flexible and weight-saving film with a low filler content that is suitable for use in lithium-sulphur batteries. The film should have a thickness of 0.1-0.3 mm and achieve a minimum through-plane conductivity of 1 S m^{-1} in order to be flexible and bendable.

In the first phase of the investigation, a series of tests was carried out based on SEBS compound and polypropylene compound (EEE) in order to develop promising mixing ratios.

The test series comprised various mixing ratios of SEBS compound and EEE, whereby the processability was improved by adding a silicone lubricant. The results showed that the optimum mixture was a ratio of SEBS to EEE of 30:70, with an in-plane conductivity of around 346 S m^{-1} and a through-plane conductivity of around 60 S m^{-1}, well above the project requirements.

In the second phase, the optimum compound candidate (SEBS/EEE+GM 30/70+20) was tested in a calendering process. The compound was applied to heated rollers, melted and processed into a film with an initial thickness of 0.21 mm. Through further parameter modifications, the aim is to reduce the thickness to 0.1 mm in order to fulfil the requirements for flexible battery electrodes.

The development of a bipolar structure of the lithium-sulphur battery has the potential to create significant improvements in various areas of application. In particular, these batteries could further advance the field of electromobility due to their low weight and high energy density. In terms of large-scale energy storage, this type of battery can also help to increase efficiency on a larger scale and reduce costs in the long term.

The "BiPoLiS" project represents an important advance in lithium-sulphur battery technology. By combining classic battery architecture and the bi polar structure, this innovative combination is intended to meet the currently limiting challenges. The results make a significant contribution to the further development and industrialisation of the lithium-sulphur battery.

For future investigations and developments in the field of film extrusion, the process parameters could be optimised in order to further reduce the film thickness and simultaneously optimise the electrical conductivity and mechanical properties. The use of electrical conductor foils based on polymer-graphite compounds in lithium-sulphur batteries could significantly improve the performance and service life of these energy storage devices. Furthermore, investigations into the long-term stability and scalability of production are required in order to exploit the potential of this new material and realise its commercial application in bipolar battery designs.

6. AVAILABILITY OF BIPOLAR PLATES

All graphite compound bipolar plates and conductor foils are produced and available at Whitecell Eisenhuth

CONFLICT OF INTERESTS

None.

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