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Flexible and Conducting Metal-Fabric Composites Using the Flame Spray Process for the Production of Li-Ion Batteries

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The wire flame spray process was used to produce electrically conductive and flexible Al coatings onto diverse textile fabrics. The investigation studied the influence of the spraying parameters and fabric materials on the electrical conductivity of the metal-fabric composites. Furthermore, this study showed that the production of flexible Li-ion batteries having good electrical properties based on the use of such flame-sprayed aluminium cathode current collectors is viable. Results show that a coating quantity threshold of about 20 mg/cm^2 exists to obtain a sufficient electrical surface conductivity for a commercial use of the produced metal-fabric composites. An excellent electrical surface conductivity of the composites (about 500 S_A) could be achieved through an adequate optimization of the spraying parameters. This conductivity increase enabled a reduction of the coating quantity and thus the flexibility of the fabric materials is better conserved, rendering the use of such composites for flexible batteries even more interesting. This study showed that the production of electrically conductive and flexible metal-fabric composites having sufficient electrical conductivity for the manufacture of flexible Li ions batteries is possible. This new method of producing such batteries represents an alternative to other chemically based processes which are hazardous to the environment because of their chemical nature.

Keywords aluminium, electrically conductive coatings, flame spraying, textile substrates

1. Introduction

The development of new energy storage methods is a priority research topic especially in transportation and combines multiple scientific disciplines such as physics and chemistry. Some of these researches are concentrating themselves on optimizing actual energy storage methods (Ref 1-4), while others are focusing themselves on the development of new storage possibilities (Ref 5-8), such as those described in the present study.

The primarily aim of the present study is to show that the production of flexible Li-ion batteries based on the wire flame spraying of fabric textiles is viable. Owing to their flexibility, these batteries could be used in applications for which the available space for the battery implementation is importantly restricted, in applications for which the battery should conform itself to a certain complex geometry or in applications which consist of integrating the battery or some electrically conductive coatings into a piece of clothing (Ref 9-14), just to name a few.

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The structures of such Li-ion batteries are based on the use of an Al-based cathode current collector along with a copper-based anode current collector, both having an active coating and being separated by an appropriate electrolyte substance, as shown in Fig. 1. Several production routes exist for producing the Al- and Cu-based cathode and anode current collectors. Such routes consist of using either wet chemical processes such as plating or physical processes such as PVD (Ref 15, 16) or sputtering (Ref 17). However, these routes are either environmentally detrimental or hazardous (wet chemical processes) or produces coating on fabric textiles which tend to be brittle and not tear resistant. Therefore, an alternative route for producing such electrically conductive coatings is needed, and the flame spray process was therefore chosen for this study. For the present paper, only the Al-based cathode current collector will be produced using the flame spray process, whereas the Cu-based anode current collector will still be produced using conventional techniques. For the future, it is planned to fabricate flexible batteries entirely consisting of flame-sprayed cathode and anode current collectors.

Flame spraying is a well-known and established process in the thermal spray industry for producing coatings for several types of applications, such as wear and/or corrosion resistance and refurbishment of worn parts, just to name a few. Since the spray jet temperature involved in flame spraying (through combustion of acetylene with oxygen) is relatively low (about 3000 °C), flame spraying may only be used for spraying metallic materials or alloys. However, this temperature limitation represents an important advantage with regard to other thermal spray processes such as plasma (for which the jet temperature can

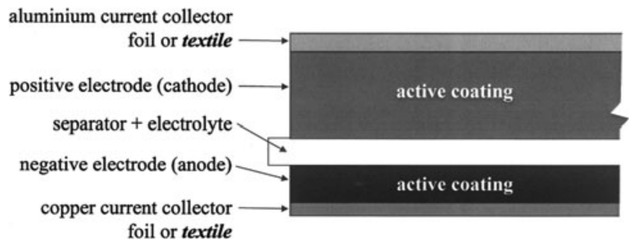

Fig. 1 Typical structure of a Li-ion battery

reach up to 20000 K) for producing coatings on heat-sensitive substrates such as textile fabrics. HVOF and cold-gas spray processes could also be alternatives to flame spray, since their jet temperatures are quite similar (HVOF) or importantly lower (cold gas) than the flame spray process. Cold spray was previously used to produce electrically conducting coatings on polymer substrates (Ref 18, 19). However, for both processes (HVOF and cold gas), the extremely high particle velocities reached may be detrimental to the textile substrates. Flame spray was even being used to produce conductor tracks or simple circuit boards on plastic substrates (Ref 20, 21), but up to now, only a couple of studies were focused on the production of ceramic coatings on fabric materials using the flame spray process (Ref 22, 23). Therefore, flame spray remains the best alternative for producing electrically conductive coating onto textile fabrics.

2. Materials and Experimental Procedure

Aluminum wire having a diameter of 3.2 mm (product number 50.11.2 from GTV-mbH, Luckenbach, Germany) was used to produce coatings onto various textile materials, using a GTV 12E wire flame spray gun (GTV-mbH, Luckenbach, Germany). Aluminum powders were not taken into account since it had been reported earlier that the use of wires produced coatings having higher specific electrical conductivities than those obtained while using powders (Ref 24, 25). The spray gun was mounted on a six-axis robot (ABB, Model IRB2400/S4C, Västerås, Sweden), while the fabric substrate was maintained in a fixed position during the spray process.

The pre-cut textile substrates were fixed in a specimen holder having dimensions of approximately 20 cm × 30 cm. As shown in Fig. 2, the specimen holder consisted of three parts: two aluminium frames having a thickness of 3 mm between which was inserted a steel mesh for maintaining the textile substrate in an adequate position and for eliminating any deformation of the substrate due to the particles and gas velocity during the spray process. The 3-mm thickness was chosen to avoid any heat-induced deformations of the aluminium frames during the spray process. Apart from cutting the textiles into dimensions which fitted the aluminum frame specimen holder, no further surface preparation was performed on the textile substrates prior to the spray process.

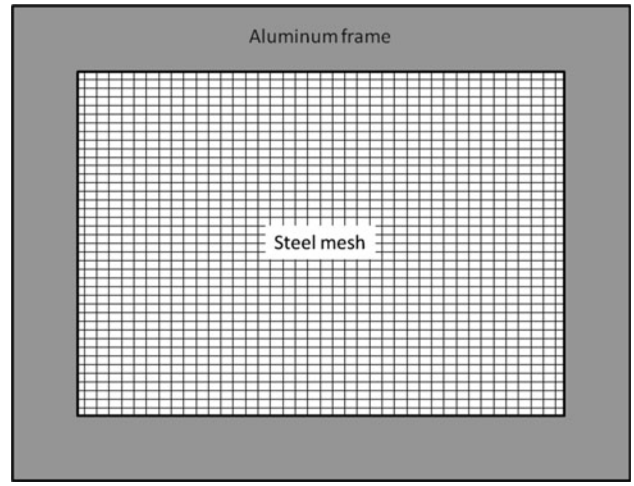

Fig. 2 Specimen holder used for spraying onto fabric textile substrates

In order to shield the heat-sensitive textile substrates from the direct exposure to the flame of the wire spray gun during the coating process, two air flow concentrators (EPUTEC, model 120022, Kaufering, Germany) were mounted on the robot's arm and which allowed projecting concentrated ambient air onto the substrate surface. The distance between these air concentrators and the substrate surface was always equal to the standoff distance. Along with the use of such air concentrators, an optimization of the spray distance was also performed (and presented later) to reduce and eliminate any heat damage that could occur on the textile substrates.

Since for coated textiles, the coating thickness may be irrelevant, especially for textiles having an open-mesh structure, the specific coating weight was used to determine the amount of material deposited on the substrate. This specific coating weight was determined by weighing the coated substrates after the spray process, subtracting the textile weight, and dividing the result by the coating area:

$$\text{specific coating weight}\left[\text{mg/cm}^2\right] = \frac{\text{weight coated substrate} - \text{weight uncoated substrate}}{\text{coating area}}.$$

Scanning electronic microscopy was used to qualitatively analyze the structure of the coated textiles' surfaces and cross sections to observe and determine the extent of the coverage of the fabric textile fibers by the sprayed particles.

The specific surface electrical conductivities (S_A) of the different coated fabrics were measured using a self-developed 4-points surface conductivity measurement setup, as shown in Fig. 3, along with a commercial high-end multimeter (Keithley, model 2010). The units of the measured values (Siemens) are identified as S_A, where the subscript "A" is added to remind the reader that these are surface-relevant values. Further details on the exact procedure used for the electrical conductivity measurements are reported elsewhere (Ref 24).

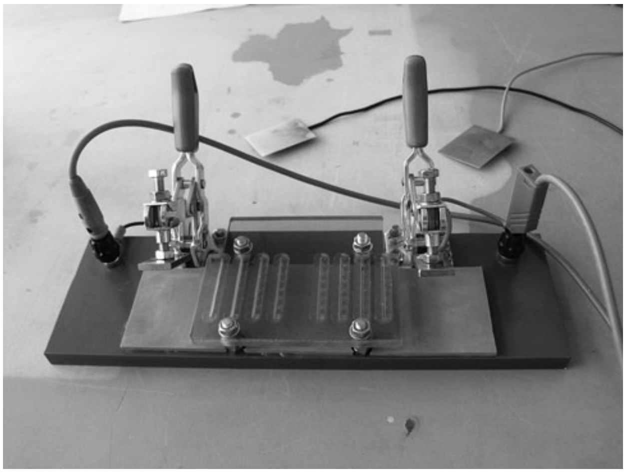

Fig. 3 Setup for the 4-points surface conductivity measurements

Table 1 Wire flame spray parameters used for the production of metal-fabric composites

Wire flame spray parameter	Value
Acetylene flow rate, l/min	~18
Oxygen flow rate, l/min	~30
Optimized standoff distance, mm	300
Step size, mm	13
Wire speed, m/min	2.5
Robot linear speed, m/min	60

Since the aim of this study is to demonstrate the possibility of producing flexible batteries out of aluminum-coated textiles using the flame spray process, the reader is referred to the studies published earlier by the author to have a much profound insight on the optimization procedure of the spray parameters (Ref 24, 25). For clarity purposes, only a brief overview of the spray parameters and their optimization and of the textile materials used will be given here.

Table 1 lists the main wire flame spray parameters used for the production of the Al-coated textile substrates. The acetylene and oxygen flow rates are the standard rates suggested by the manufacturer for the Al wire. In order to perform an easy and quick optimization of the spray parameters, the flow rates were not optimized, but only the standoff distance was.

Since the deposition efficiency of flame-sprayed Al coatings irrespective of the substrate materials used is strongly dependent on the standoff distance, this spray distance was optimized to obtain good deposition efficiency without incurring any heat-induced fabric degradations. This optimization consisted of performing several spray experiments onto the selected fabric materials, beginning with a relatively large distance (several hundred millimeters) and reducing this distance until heat-induced fabric degradations could be observed through the naked eye.

Several fabric materials were tested in this study but for clarity purposes, the four most interesting textile substrates with regards to the obtained properties will be presented. The first two fabrics (having the denomination 5248 and 5911) were made of polyester and had a densely woven mesh pattern. A third fabric, named C2010 was also made of polyester and had an open-mesh pattern. The fourth fabric, named "XX" which was Kevlar-based had a significantly loose mesh structure. For all experiments, one-sided and two-sided coated textile substrates were produced to study the influence of the overall coating of the substrate on its conductivity.

Besides the whole-coated textile samples (area of approximately 20 cm × 30 cm), simple circuits were also fabricated onto selected textile materials using an appropriate contact masking method to evaluate the feasibility of producing such circuits using the flame spray process. The developed masks, as shown for example in Fig. 4, were made of an aluminum sheet having a thickness of 3 mm which was laser cut for the desired configuration. Again, the 3-mm thickness was chosen to avoid any heat-induced deformations of the mask during the spray process. Different mask geometries were tested, but for clarity purposes, only the example shown in Fig. 4 will be discussed in the present paper. The mask was inserted between the textile substrate and the front aluminum frame using the substrate holder (Fig. 2). The optimal spray parameters were used for producing the simple circuit boards. It is worth noting that the textile circuit board has different gap sizes from 4 mm down to 1.5 mm.

For the characterization of the flexible batteries having an anode current collector consisting of a flame-sprayed Al-coated fabric, the overall capacity of the manufactured batteries along with their charging and discharging behaviors were studied and compared with button cells also made with flame-sprayed Al anode current collectors and batteries made with Al foils. The test procedure consisted of repeated charge/discharge cycles (charge: constant current at 4.2 V followed by constant voltage until current <10 mA; discharge: constant current until 3.2 V) were done at different currents, starting with a current value where the theoretical capacity (C_{th}) should be reached in 10 h (C-rate = 0.1). The maximum measured capacity under these conditions was set as the nominal value C_N (100%). After 4-5 cycles, the current was increased to higher C-rates (0.2-1) for another 4-5 cycles per setting. This procedure results in capacity loss profiles which consist of reversible and irreversible contributions. This test procedure is schematically illustrated in Fig. 5.

3. Results and Discussions

Like mentioned earlier in the previous section, only the spray distance was optimized by varying it until an onset of heat-induced fabric degradations on the fabric substrates could be observed. Furthermore, the specific surface electrical conductivity of the produced Al flame-sprayed substrate was measured. Figure 6 shows the electrical conductivities of the 5248 fabric substrates produced with different standoff distances. It is worth noting that this

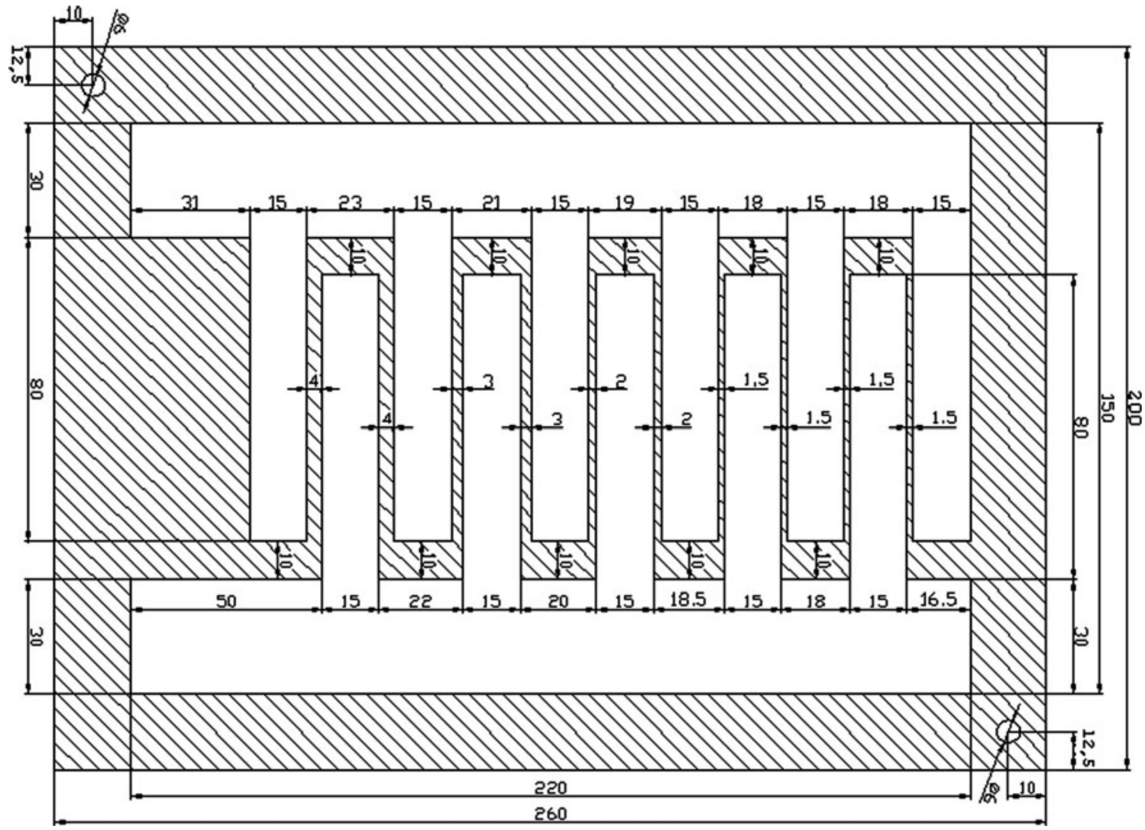

Fig. 4 Example of mask used for the production of simple circuit boards

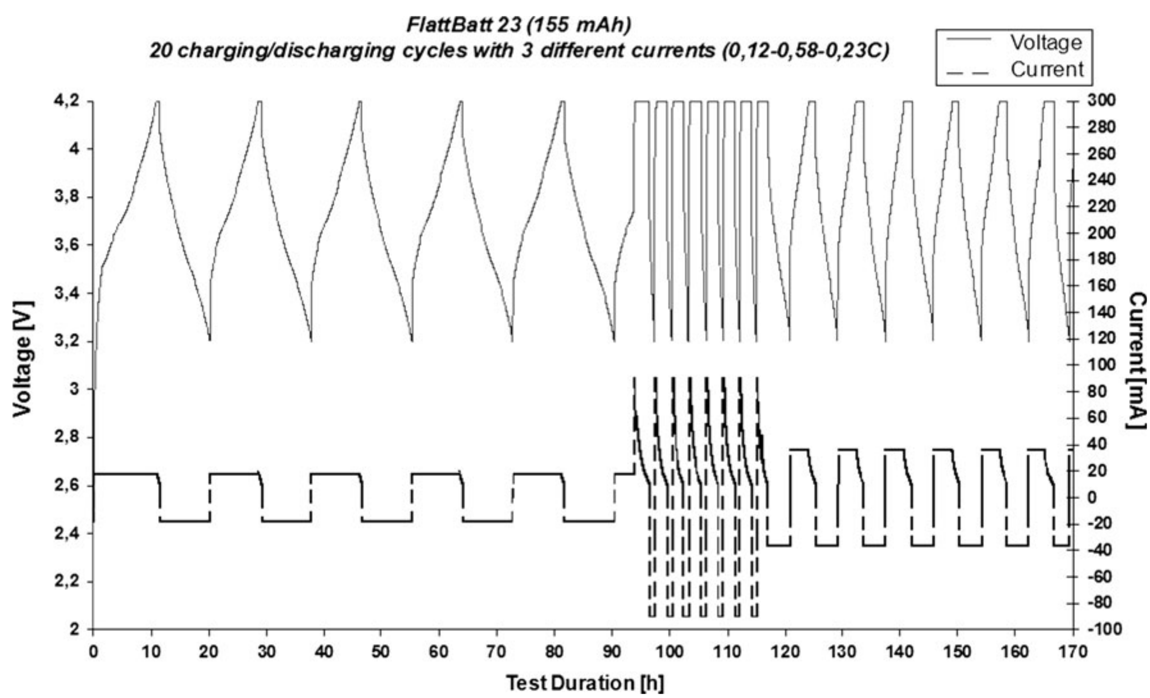

Fig. 5 20 charge/discharge cycles with three different current values for FlatBatt 23 (155 mAh) with a cathode current collector made from Al-coated 5911 fabric material

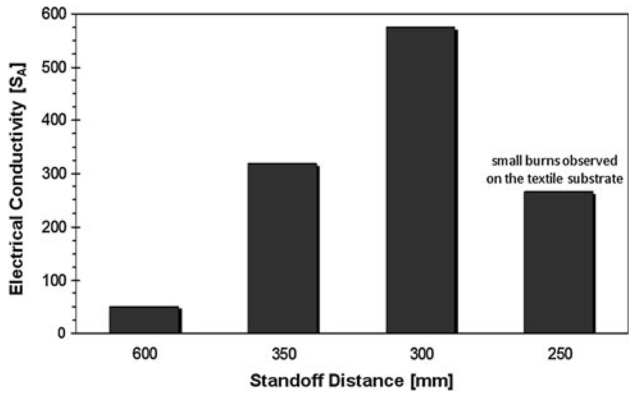

Fig. 6 Influence of the standoff distance on the electrical conductivity of the 5248 fabric substrates

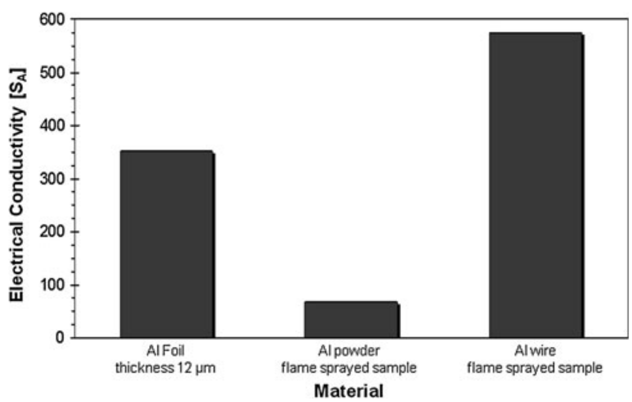

Fig. 7 Comparison of the specific surface electrical conductivities of an Al foil, a powder flame-sprayed Al coating, and a wire flame-sprayed Al coating, each of which applied onto the 5248 fabric material

optimization resulted in similar values for the optimized standoff distance as shown in Fig. 6 for the other three fabric materials, namely the 5911, C2010, and XX fabrics. As the standoff distance is reduced from 600 to 300 mm, the conductivity values are increasing from approximately 50 S_A up to 575 S_A. By reducing further the standoff distance to 250 mm, small heat-induced damages were observed on the fabric material rendering this spray distance unusable for producing adequate metallic coating-fabric composites. Furthermore, these burns did also influence the electrical conductivity value of the produced Al coating, since this value dropped down to approximately to 250 S_A, as shown in Fig. 6.

For comparison purposes, the specific surface conductivities of an Al foil having a thickness of 12 µm and of a powder flame-sprayed Al coating were compared to the values of the wire flame-sprayed Al coating, as shown in Fig. 7. It is worth noting that the optimum spray parameters for the powder and the wire flame-sprayed coatings were used, to compare the best achievable electrical conductivity of both flame-sprayed coatings. It is obvious from Fig. 7 that the use of wires instead of powders for the flame spray process produces significantly more conductive Al coatings onto fabric substrates, and this is the reason why no further experiments were performed with the powder flame process. It is believed that the conductivity of the wire flame-sprayed coatings is higher than the powder flame-sprayed ones because of the slightly higher particle velocity of the wire spray process. This slightly higher velocity should produce coatings having a reduced porosity in comparison with the powder flame-sprayed coatings. Furthermore, this higher particle velocity produces a slightly higher peening effect on the coating and therefore enables a better contact between the different splats by breaking partially the oxide present at the splats surface. The combination of a reduced porosity and a better contact between the splats are believed to be the major causes of the higher electrical conductivity of the wire flame-sprayed coatings. Furthermore, the wire flame-sprayed coating possesses a significantly higher conductivity value than a commercially available Al foil (575 S_A versus 350 S_A). It is worth noting that the major cause of this conductivity differences between the Al foil and the flame-sprayed coatings may be due to their different thicknesses. However, if one normalizes the conductivity with regard of the thickness, then the Al foil would have a significantly higher electrical conductivity than the flame-sprayed coatings because the foil does not possess any pores or any oxide inclusions. However, the purpose of this comparison is to show that the wire flame-sprayed coatings possess a higher electrical conductivity than a commercially available 12-µm-thick Al foil, which was thought, at the beginning of the present study, to be also a viable solution to produce such flexible current collectors.

Figure 8 shows the aluminium-coated surfaces of the four fabric materials used. The open-mesh pattern of fabric C2010 and the loose mesh pattern of fabric XX (Fig. 8a, b) could still be distinguished after the spraying process. For the two other fabric materials (5248 and 5911, Fig. 8c, d), the coated surfaces are shown in Fig. 8(c) and (d), and the fabric mesh structures are not so obvious to observe as for the first two fabric materials.

The influence of the fabric material onto the specific surface conductivity values is shown in Fig. 9 for the fabrics 5248, 5911, and XX. The influence of the substrate nature on the electrical conductivity of the sprayed coatings with coating amount being 14 mg/cm^2 is thought to be neglible because the fabrics are extremely softer than the impinging Al particles. It is worth noting that the coating amount values for double-sided fabric materials were divided by 2 to obtain the coating amount per side and to enable the comparison with the single-sided 5911 fabric. As shown in Fig. 9, a double-sided fabric possesses a significantly higher surface conductivity (factor of about 100) than a single-sided fabric (comparison of double-sided with single-sided 5911 fabric) for the same coating amount. This is due to the interconnectivity or through-contacting of both Al-coated sides of the fabric material. For example, to obtain a single-sided 5911 coated fabric having an electrical conductivity of approximately 100 S_A, the coating amount should be around 25 mg Al/cm^2, whereas for a double-sided 5911 fabric, the coating

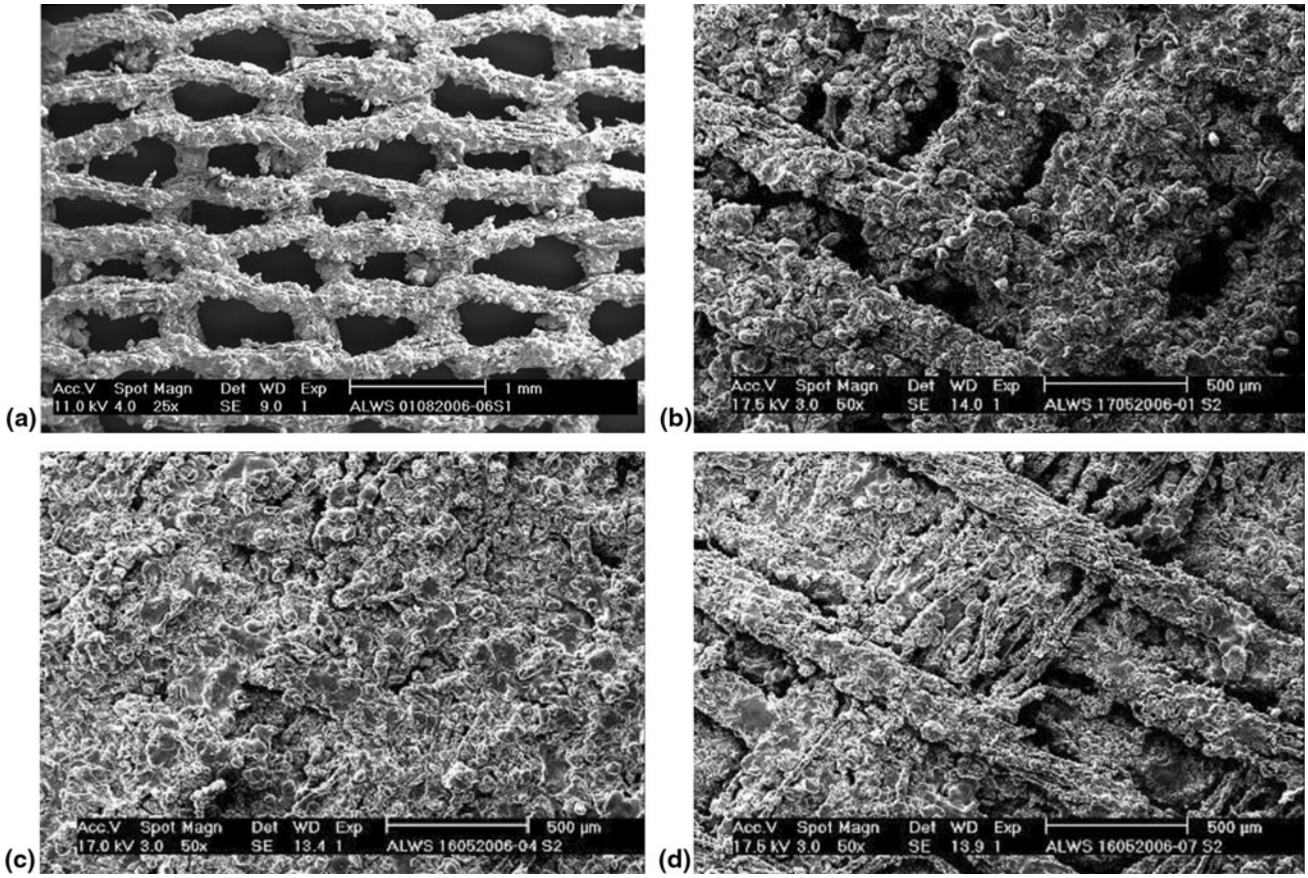

Fig. 8 Pictures showing the Al-coated surface of the different fabric materials used as substrate: (a) fabric C2010, (b) fabric XX, (c) fabric 5248, and (d) fabric 5911

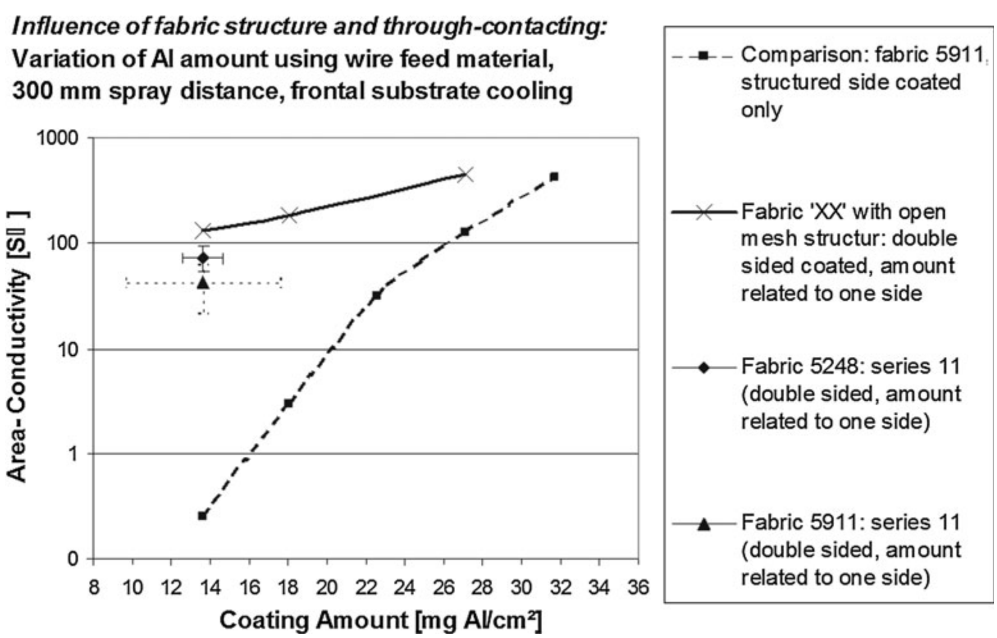

Fig. 9 Influence of fabric type, structure, number of coated fabric sides, and coating amount onto the specific electrical conductivity

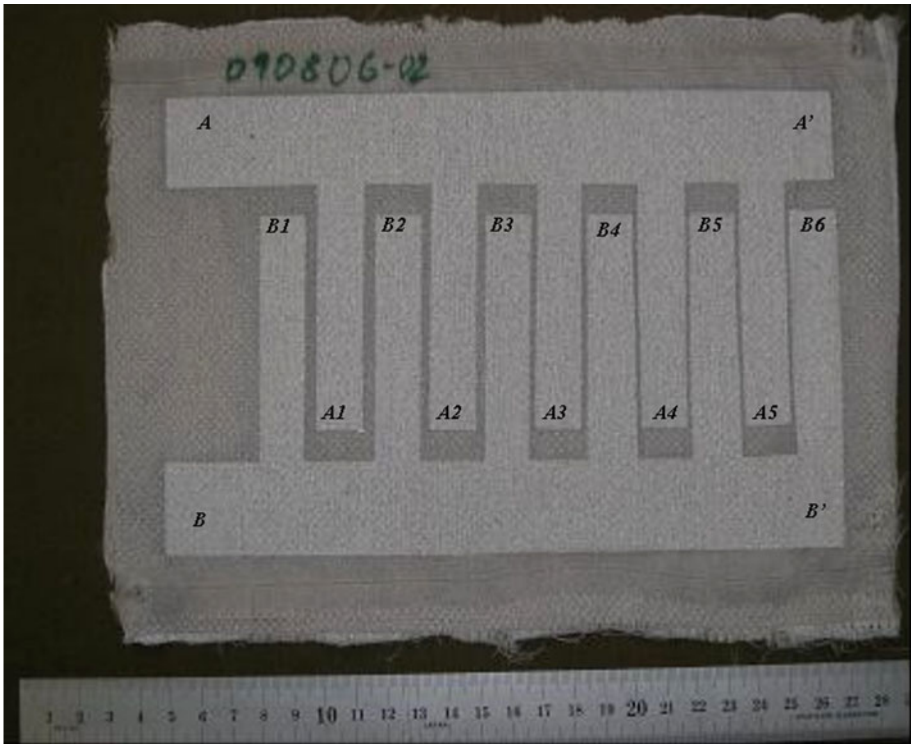

Fig. 10 Flame-sprayed textile Al-circuit board along with the conductivity measurements points

amount is around 15 mg Al/cm^2. The higher coating amount associated with the single-sided fabric material decreases significantly the flexibility of the coated fabric. Thus, the use of double-sided fabrics has the advantage of producing more flexible fabric materials for a certain electrical conductivity.

Owing to the open-mesh structure of the C2010 fabric material, it was very difficult to measure reliably any electrical conductivity value using the developed measurement setup used in this study. Therefore, no conductivity values are presented herein. However, owing to the 3D nature of the Al-coated C2010 fabric, which is believed to enhance the overall capacity of Li-ion battery, cathode current collectors using this open-mesh-structured fabric were produced. The properties of these batteries along with batteries made with the other fabric materials will be presented later.

An overall number of seven samples of textile circuit boards having different gap sizes from 4 mm down to 1.5 mm were prepared using the mask shown in Fig. 4 and two of these samples were double-sided. Apart from one, all the other samples had well-separated tracks without any measurable electrical contact, and three of them were sufficiently conducting (measured resistance below 1 Ω from main contact to the end of a "finger": Fig. 10 shows the resistance measuring points on a coated fabric) for being subsequently tested with mounted LEDs. These results show that simple circuit boards possessing sufficient conductivities could be produced using the wire flame spray process.

Figure 11 shows typical cross sections of single-sided and double-sided Al-coated 5911 fabric materials. As one may observe, the fabric fibers can be easily seen, and the coatings for both the single-sided and for the double-sided-coated fabrics have thicknesses in the range from approximately 100 to 200 μm, and the coating surfaces possess a high roughness. The flame-sprayed Al coating is significantly porous, but as will be shown later, the electrical conductivity is nonetheless still high. It is obvious that a further optimization with regard to the coating porosity would produce coated fabric materials with higher conductivity values; however, since this study was to demonstrate the feasibility of producing flexible electric conductive fabrics and flexible Li-ion batteries, this further optimization will not be presented or discussed in this paper.

Flexible Li-ion batteries based on the use of cathode current collectors made of flame-sprayed Al coatings onto fabric materials were manufactured on the principle shown in Fig. 1. Figure 12 shows the actual typical layout of such a cathode current collector: a fabric material is coated with a flame-sprayed Al coating, and an active coating (cathode material) was applied onto this Al coating. This whole cathode was then mounted with a Cu-based anode to produce a whole Li-ion battery as shown schematically in Fig. 1.

The electrical performance characterization of Li-ion batteries consisting of a flame-sprayed Al-coated fabric cathode current collector was performed using the procedure described earlier and presented in Fig. 5. The

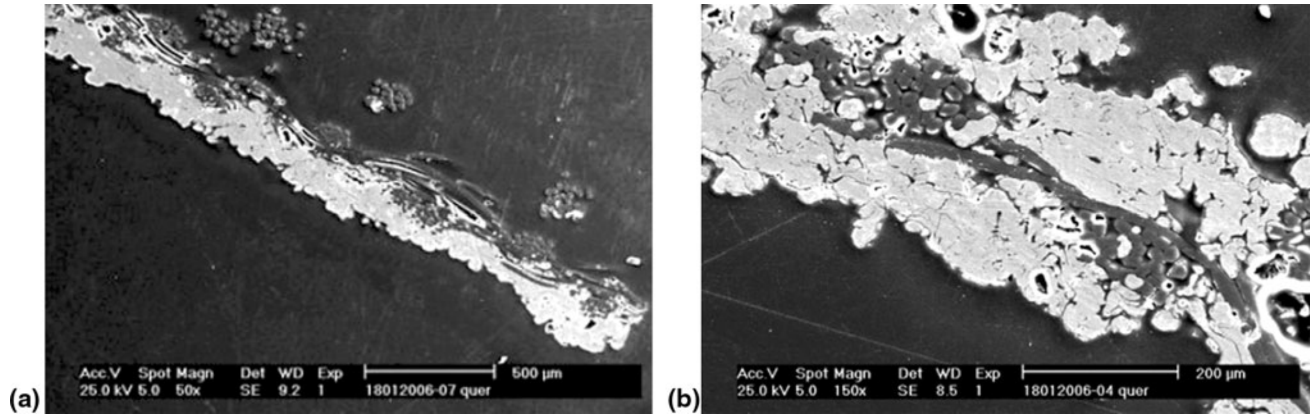

Fig. 11 Cross sections of flame-sprayed Al-coatings on (a) single-sided and (b) double-sided 5911 fabric material

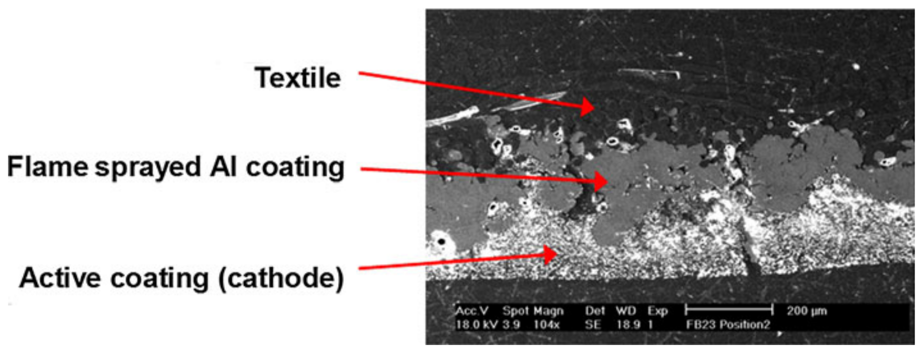

Fig. 12 Cross section of the flame-sprayed Al cathode current collector of a typical Li-ion battery

Table 2 Properties and capacity of flame-sprayed cathode current collectors in comparison with Al foil and PVD-aluminized polyester fabrics

			Cathode thickness, µm		Capacity, mAh	
Name	**Battery type**	**Cathode**	**Active coating**	**Total**	**/cm^2**	**Total**
FB23	FlatBatt	Al flame sprayed	150 (80-200)	~500	3.88	155
CCA3	Button cell	Al flame sprayed	115 (80-200)	~250	2.80	5.6
F1	Button cell	Al foil	40	55	1.25	2.5
F3	Button cell	Al foil	70	85	2.30	4.6
R1	Button cell	Al PVD	50-120	120	2.20	4.4

flame-sprayed cathode current collectors were compared with other solutions like plain Al-foil and thin PVD-coated polyester fabric. Two types of battery forms were produced and tested: flat battery (FlatBatt) and button coin cells. The summary of the FlatBatt 23 data along with the coin cell results are given in Table 2 and shown Fig. 13. The results listed in Table 2 show clearly that the total capacity of flame-sprayed cathode current collectors (in coin cell or FlatBatt form) is significantly higher than the capacity of collectors produced using other processes such as an Al foil or PVD. Furthermore, as shown in Fig. 13, these test results indicate that the flame-sprayed current collectors may be at least as good as foil collectors when it comes to dynamic capacity retention of thick active cathode layers.

All batteries listed in Table 3 were tested following the same test procedure described earlier and are shown schematically in Fig. 5. By using different fabrics, general and specific fabric-related trends were observed. In general, flat batteries have higher total capacity than their button cells counterparts (FB36, FB37, and FB38 in comparison with FBC002). Flat batteries made of flame-sprayed Al-coated cathode current collectors (all FB-numbers) all possess higher total capacity than collectors made of Al foils (FB43 and FB44). With regard to the fabric material, the fabric C2010 has the highest capacity per surface area (FB42 with 5.10 mAh/cm^2). In summary, the total battery capacity could be increased to 400 mAh, and the charge density to 5.1 mAh/cm^2, when using different fabric materials for the cathode current collectors.

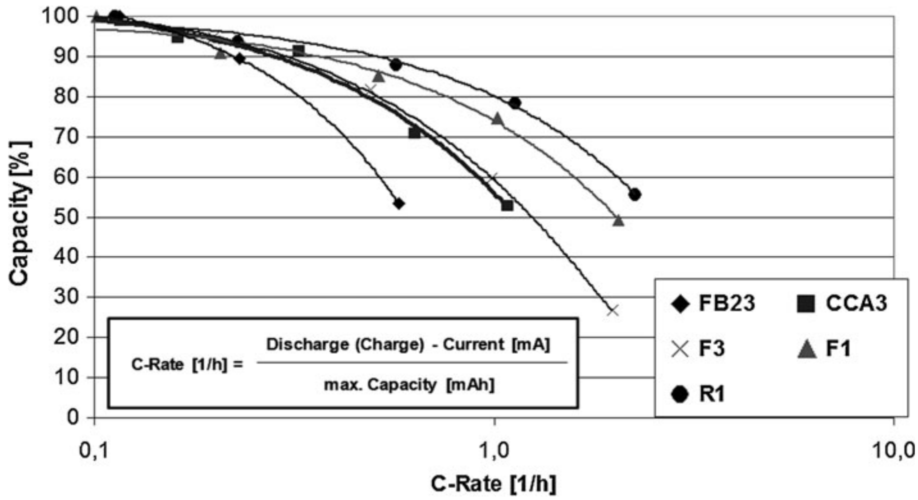

Fig. 13 Performance comparison: Al-flame-sprayed cathode current collectors in comparison with Al foil and PVD-aluminized polyester fabrics

Table 3 Summary of the test batteries results

Name	Battery type	Cathode collector fabric/Al-layer/side	Capacity, mAh /cm^2	Capacity, mAh Total	C_N/C_{Th} [%]
CCA 3	Button cell	5248/1/1	2.80	5.6	89
FBCC02	Button cell	XX/2/2	3.80	7.6	98
FB23	FlatBatt	5911/1/rough	3.88	155	87
FB27	FlatBatt	5911/1/smooth	2.83	113	91
FB35	FlatBatt	XX/2/1	2.20	88	84
FB36	FlatBatt	XX/2/2	2.83	113	84
FB37	FlatBatt	XX/2/2	2.25	90	83
FB38	FlatBatt	XX/2/2	3.03	121	84
FB39	FlatBatt	Al foil	2.05	82	88
FB40	FlatBatt large	XX/2/2	2.60	416	81
FB41	FlatBatt large	XX/2/2	2.52	403	78
FB42	FlatBatt	C2010/2/2	5.10	204	83
FB43	FlatBatt	Al foil	1.25	50	73
FB44	FlatBatt	Al foil	2.05	82	80
FB45	FlatBatt (three electrodes)	C2010/2/2	3.90	312	82

Table 4 General influences of different Al-coated cathode current collector fabrics on the cathode properties

Fabric style	Capacity	Flexibility	Weight
Densely woven			
Smooth (5248)	−−	+	−−
Rough (5911)	+	++	−
Loose (XX)	+	+	+
Open mesh (C2910)	++	−−	++

A benchmark for a good FlatBatt is FB38, which has values of 121 mAh and 3.03 mAh/cm^2. FB35 and FB37 are of somewhat less quality despite a lower power density, which may be due to variations during the battery manufacturing. The larger batteries FB40 and FB41 (electrode area increased four times to 160 cm^2) correspond well with the comparable smaller FB36. The battery performances using the open-mesh fabrics (FB42: two electrodes set-up; FB45: sandwich of two cathodes with a double-sided anode in between) show an interesting potential for the future development of flexible Li-ion batteries.

An empirical judgement of all the experiences performed so far in the manufacturing of cathode current collectors out of different fabrics is given in Table 4. The highest battery capacity may be obtained when using an open-mesh fabric like the C2010, but the overall flexibility is not so good. A good compromise regarding the battery capacity, its flexibility, and its weight lies in the use of a loose mesh fabric, like the fabric XX, for which all the three properties are good (but not excellent). From Table 4, one may observe that depending on the property of the flexible Li-ion-based battery which is aimed as important for the specified application, one has to choose the adequate fabric material structure to obtain an optimized and tailored battery related to this application.

The use of such flexible Li-ion-based batteries in combination with flexible textile simple circuit boards

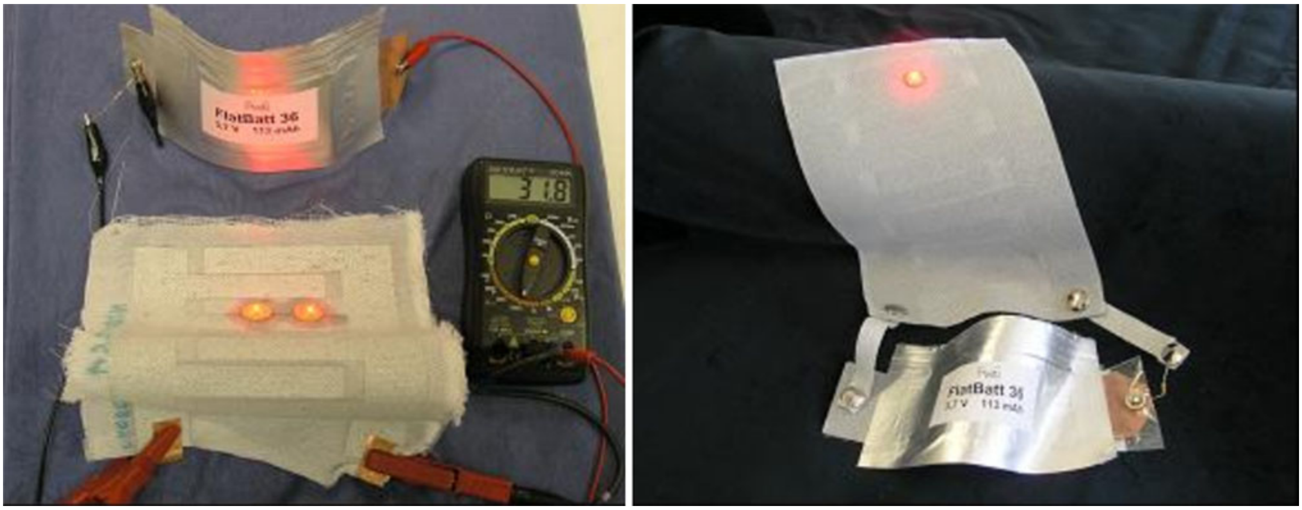

Fig. 14 LEDs mounted on flame-sprayed flexible circuit boards powered by a flexible battery (FB36)

(already shown in Fig. 10) is demonstrated in Fig. 14. LEDs mounted onto a simple circuit board are alimented by a flexible battery (FB36) connected to this board. Figure 14 demonstrates without any doubt that flexible Li-ion batteries and simple circuit boards may be produced using flame-sprayed Al-coating on fabric materials.

4. Conclusions

In this study, the wire flame-sprayed process was successfully used to produce electrically conductive and flexible Al coatings onto diverse textile fabrics. Furthermore, this study showed that the production of electrically conductive simple circuit boards and flexible Li-ion-based batteries consisting of flame-sprayed aluminium cathode current collectors having sufficient electrical conductivity for any specified application is possible and viable.

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