Investigation of Mechanical and Electrochemical Performance of Multifunctional Carbon-Fiber Reinforced Polymer Composites for Electrical Energy Storage Applications

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Summary: Concept of structural supercapacitors, based on carbon fiber reinforced polymer composites, has been introduced that can act as a structural load bearing component as well as an electrical energy storing device simultaneously. This multifunctional carbon fiber reinforced structural supercapacitors are fabricated by using carbon fiber and glass fiber/filter paper as reinforcements and cross-linked polymer electrolyte as a matrix. Carbon fiber mats also simultaneously serve the role of electrodes in addition to reinforcements whereas the glass fiber mat/filter paper also acts as an insulator to avoid the short-circuiting of the carbon fiber electrodes. A polymer epoxy matrix is modified by introducing ions within the cross-linked structure in order to develop an optimized polymer electrolyte. Flexural tests of structural supercapacitor are conducted to evaluate the structural performance while charge/discharge tests are conducted to evaluate the electrochemical performance. Multifunctional structural supercapacitors are tested mechanically as well as electrochemically. A structural supercapacitor is fabricated showing simultaneously an energy density of 0.11 mWh m^{-3}, a specific capacitance of 0.8 $mF.cm^{-3}$ and a flexural modulus of 26.6 GPa simultaneously.

Keywords: Multifunctional composites; Polymer electrolytes; Structural supercapacitors; Flexural properties; Electrical energy storage.

Introduction

In the recent past, a large amount of work has been done for the development of multifunctional structural composites [1; 2]. Structural functions contain mechanical properties like strength, stiffness, toughness, and damping [3], while non-structural functions include electrical and/or thermal conductivity [4], sensing and actuation [5], energy harvesting/storage [6-11], self-healing capability [12], electromagnetic interference (EMI) shielding [1], recyclability and biodegradability. In particular, structural supercapacitors have gained a tremendous interest as they simultaneously offer electrical energy storage as well as bear mechanical loads. Supercapacitors, which are also known as electric double layer capacitors (EDLC) or ultracapacitors, are capacitors with capacitance values greater than any other capacitor type, available today. Capacitance values reaching up to 8 $F.cm^{-3}$ in a single standard case size are commercially available [13]. The first general use of ultracapacitors was low-power, low-energy, and long-life back-up power sources for consumer electronics such as video cassette recording (VCR). Today, ultracapacitors are used for many different industrial applications [15].

In a supercapacitor, an electrolytic double layer is created which increases its electrochemical performance [15]. When two pieces of carbon electrodes are immersed in an electrolyte, they form an amazingly effective double capacitor connected in a series. Structural supercapacitors are usually made with carbon, separating them with a thin separator, and impregnating the layup stack with an electrolyte. The fact, which makes these supercapacitors a high source of energy, is the use of carbon technology. The electrochemical capacitor is made with a carbonized porous material as an electrode, which has a large surface area due to the cavities. The thickness of the dielectric layer equals the distance between the electrodes. For having high capacitance, the insulator is kept very thin [15]. Supercapacitors have high capacitive energy density and can be used for applications that were reserved for only batteries in the past [15]. The most important advantage that supercapacitors have over batteries, is their ability to be charged and discharged continuously without degrading for large number of cycles. Thus batteries and supercapacitors can be used in conjunction with each other as hybrid energy storage devices [15]. The structural supercapacitor technology is relatively new and is constantly evolving [6-8].

In this study, structural supercapacitors have been fabricated using carbon fibers as electrodes and

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reinforcements, glass fibers/ filter paper as separator and reinforcements and vinyl ester resin asan electrolyte and polymer matrix. The effect of different salts, as well as separators on the electrochemical and mechanical performance of structural supercapacitors, has been explored.

Materials

Materials used in the formation of multifunctional structural supercapacitor were, carbon fiber (plain weave, PAN-based, 200 $m^2.g^{-1}$ areal density, Easy Composites, UK), filter paper (Grade 1, Whatman, UK), glass fiber (plain weave, Tissa Glasweberei AG, Oberkulm, Switzerland), Tissue fiber (chemical resistant tissue C-glass Nexus veil, France). For the formation of electrolyte, lithium perchlorate (99.9% purity, battery grade), potassium perchlorate (99.9% purity), zinc chloride (99% purity) and acetonitrile were purchased from Sigma Aldrich, UK. Vinyl ester resin (Hetron 922, Ashland USA) was also purchased.

Preparation of polymer electrolytes

For the preparation of 1M salt electrolyte solution, 10.63 g of lithium perchlorate was mixed in 100ml acetonitrile. The stirring of salt solution was done through magnetic stirrer for 30 min at 45°C for better mixing. The homogenized solution of salt and acetonitrile was stored in an airtight measuring flask to prevent it from air and moisture. Weight of various salts used in 100ml acetonitrile to prepare 1M solution is shown in Table-1.

Table-1: Weights of various salts in acetonitrile solvent used to prepare a 1M electrolytic solution. E_L_ lithium perchlorate, E_N_ sodium chloride, E_Z_ Zinc chloride, E_P_ potassium perchlorate.

Sample ID	Name of Salt	Weight (g)
E_L	**Lithium per chlorate ($LiClO_4$)**	**10.6**
E_N	**Sodium chloride (NaCl)**	**5.84**
E_Z	**Zinc chloride ($ZnCl_2$)**	**13.6**
E_P	**Potassium per chlorate ($KClO_4$)**	**13.8**

Liquid electrolyte (1M $LiClO_4$/Acetonitrile) was mixed with vinyl ester resin and after 30 min of stirring on magnetic stirrer; MEKP was added in the resin (2% by weight of vinyl ester). Other polymer electrolytes were also prepared by varying electrolyte content to 5 wt. %, 10 wt. %, 15 wt. % and 20 wt. % electrolyte.

Fabrication of Structural Supercapacitor

After the selection of the polymer matrix, reinforcement, electrolyte, and the separating medium, the supercapacitor was fabricated using "hand lay-up method". During fabrication, the first step was the preparation of glass molds by cleaning them properly and mold releasing agent was applied to them. The second step was the coating of polymer matrix on carbon fiber reinforcement with the help of paint brush. After coating one side of first carbon fiber mat, it was turned carefully and the coating was also applied on another side of the same carbon fiber mat and was placed on glass mold. Next step was the coating of polymer matrix on filter paper/glass fiber in the same way as coating was applied on carbon fiber mat, this coated filter paper was placed carefully on already coated carbon fiber mat, and a copper wire was also placed on extreme corner of each carbon fiber mat, between carbon fiber and filter paper. Another carbon fiber mat was also coated in the same way as done before and placed on coated filter paper in layer form and copper wire was also placed at another extreme edge of polymer matrix coated carbon fiber mat between filter paper and second coated carbon fiber. Remaining polymer matrix was applied on this layered form composite and with the help of paint brush. The second mold was then placed on it and the sample was placed in an oven at 80°C for 24 hours under atmospheric pressure. After 24 hours, composite samples were removed from the oven.

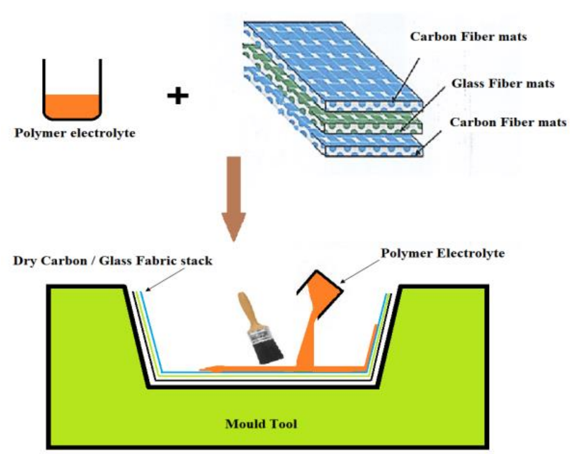

Fig. 1: Schematic for the fabrication of structural supercapacitors.

Mechanical characterization of structural supercapacitors

In three-point bending test, load is applied between two clamped ends of the sample that provides flexural strength, flexural strain, and flexural modulus. Equipment used for testing was UTM (Universal testing machine, TIRA 2810 Germany), coupled with a load cell of 10KN.

Composite samples were cut according to ASTM D7264. Span length applied was 55mm. The sample was clamped between two supporting ends of UTM and load was applied at the middle of the span. The flexural test measures the behavior of materials subjected to simple beam loading. It is also called a transverse beam test. Maximum fiber stress and maximum strain are calculated for increments of load. Results are plotted on a stress-strain diagram. Flexural strength is defined as the maximum stress and the flexural modulus is calculated from the slope of the stress vs. deflection curve in the elastic region [17].

Electrochemical Characterization of Structural Supercapacitor

The electrochemical testing (charge-discharge test) of structural supercapacitor was performed using Potentiostat/ Galvanostat (Model 263A, Princeton Applied Research, 801 South Illinois Avenue, Oakridge, Tennessee, United States). This is the most versatile model of Potentiostat/ Galvanostat as it can perform the charge/discharge when test bare portion of copper wires (used in the sample on both ends) were connected to the equipment probes. Neutralizing time set for the sample was 50 seconds. Time for charging was 150 seconds, and for discharging was 450 seconds. Charging/discharging graphs were obtained from this test.

Fiber Volume Content of Structural Supercapacitor

Quality tests of composite samples were performed in order to find out the fiber volume content of the composite in accordance with ASTM D 3171. Three coupons of each sample having exactly the same shape and dimensions (2.5 x 2.5 cm) were cut by using a cutter. Proper sizing of specimens was done before digestion. After preparing coupons, 25-mL of sulfuric acid was taken in a flask and the sample specimen was put in it and the flask was placed on a hot plate. Fumes started rising through the sample and when solution became dark brownish in color, 35-mL of hydrogen peroxide was added in the beaker. Intense fumes were raised and suddenly the solution became colorless. After removing the solution from the hot plate it was cooled down in a water bath. The solution was then filtered under vacuum. The fibers were washed with distilled water and acetone. . The specimen was placed in an oven at 100 °C for 2 hours. After drying, the fibers were taken out of the oven and their density was measured. Then the fiber content was determined accordingly.

Results and Discussion

Influence of Electrolyte Variation on Mechanical and Electrochemical Properties of Structural Supercapacitors

In order to see the effects of salt variation on the electrical and mechanical properties of the structural supercapacitor, three salts (lithium perchlorate, potassium perchlorate, and zinc chloride) were used. All these three salts were used in the same percentage with the resin i.e. in the ratio of 80:20. The electrochemical testing performed using the Potentiostat/Galvanostat (Model 263A, Princeton Applied Research, 801 South Illinois Avenue Oakridge, Tennessee, and United States). There are two main reasons behind the variations in electrical properties, first is the difference in the size of the ions of these salts. As we know in a group of the periodic table, similar charged ions increase in size from top to bottom, whereas, isoelectronic positive ions show a decrease in ionic radius from left to right, because of the increasing nuclear charge. The decreasing ionic size improves the ionic mobility within the crosslinked structure of polymer electrolyte. Lithium has the smaller ionic size than potassium but zinc has the smallest ionic size among them as it is a transition element shows different behavior from other elements. The other main reason is the conductivity of each salt. This property is mainly due to the presence of relatively loose electrons in the outermost shell of the element. The electrical conductance generally increases from top to bottom and decrease from left to right, so lithium has less electrical conductivity than potassium but the ionic size of potassium is greater than lithium which causes restriction in their flow, so it gives poor properties than lithium on the other hand although zinc has smaller size and its electrical conductivity is less than lithium and potassium. Hence, it showed that lithium salt has the best electrical properties among the three studied salts. Fig 2 shows charge/discharge plots of structural supercapacitors with varying salts.

Fig. 2 shows that the supercapacitor having lithium perchlorate as the electrolyte has the biggest charge-discharge area as compared to the other two salts because of its small size and better electric conductivity such as specific capacitance of 0.8 $mF.cm^{-3}$ and energy density of 0.473 $mWh.kg^{-1}$. On the other hand, supercapacitors having potassium perchlorate as electrolyte shows specific capacitance of $2.62x10^{-4}$ $mF.cm^{-3}$ which is less than zinc perchlorate having a specific capacitance of $1.24x10^{-2}$ $mF.cm^{-3}$ due to its bigger ionic size which causes a restriction in their flow and as result, potassium shows less electrical properties and having a less charge-discharge area. In the case of zinc salt, the area is greater than potassium although less than lithium due to its small ionic size but poor electrical properties than both lithium and potassium. The same trend is shown for the energy density of potassium and zinc having 3.64×10^{-5} $mWh.kg^{-1}$ and 1.72×10^{-3} $mWh.kg^{-1}$ respectively.

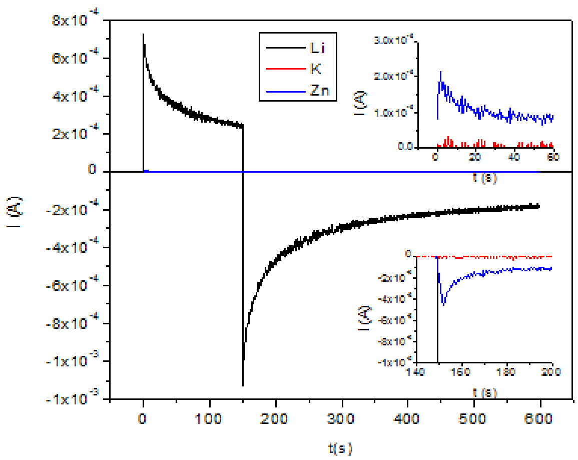

Fig. 2: Charge/Discharge plots of structural supercapacitors with varying salts.

Fig. 3 shows the stress/strain plot of structural supercapacitor using flexural three-point bending configuration with varying salts. As we know mechanical properties decrease from top to bottom so lithium has less mechanical properties than potassium while zinc is a transition metal which has very good mechanical properties and hard in nature, therefore zinc gives better properties than lithium and potassium. Although there are minute differences in their mechanical properties because small amounts of salts were used while the contribution of mechanical properties from the resin is same in all three salts, so there have very small differences in their mechanical properties. The flexural modulus of lithium, potassium, and zinc are 28.6 ± 3.62 GPa, 25.8 ± 3.32 GPa, 28.3 ± 5.82 GPa respectively. Also, zinc induces hardness due to its transition element property.

The comparison of the mechanical properties of these samples (Table-2) showed that the flexural strength and flexural modulus of the supercapacitor, having lithium perchlorate salt, were not very much less than the other two specimens having potassium perchlorate and zinc chloride. But the electrical properties of the supercapacitor having lithium perchlorate were very high as compared to the other two so it was selected as the most preferable salt to be used as an electrolyte.

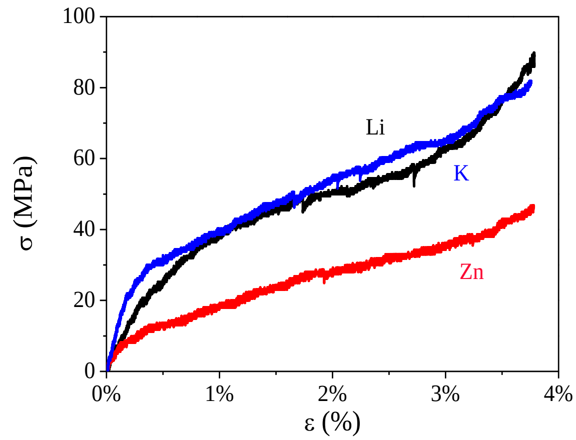

Fig. 3: Stress-strain plot of structural supercapacitors with varying salts

Fig 4 shows the optimization of various structural supercapacitor specimens with different electrolytes by plotting energy density and flexural modulus on y-axis and supercapacitors with varying electrolyte on x-axis. Fig 4 shows that lithium salt gives greater energy density as well as mechanical properties.

Influence of Varying Electrolyte Content on Mechanical and Electrochemical Properties of Structural Supercapacitors

After the selection of lithium perchlorate as the electrolyte, its concentration was varied from 5% to 20% in the matrix to study the influence of electrolyte content variation on electrical and mechanical properties of the supercapacitor. For determining the electrochemical properties of the structural supercapacitor, electric charge/discharge tests were conducted using Potentiostat/Galvanostat (Model 263A, Princeton Applied Research, 801 South Illinois Avenue Oakridge, Tennessee, United States) by varying the electrolyte content. As expected, the charging/discharging area for the 20% lithium salt-based supercapacitor was greater as compared to all the others. This happened because with the decrease in the amount of the electrolyte, fewer ions traveled in the resin and lower area of charge-discharge curve was obtained. Thus, by increasing the amount of electrolyte from 5 wt% to 20 wt%, improvements in specific capacitance, as well as energy density, was observed as highlighted in Table-3. Fig 5 shows the charge/discharge plot of structural supercapacitor with varying electrolyte content.

Table-2: Electrical and mechanical properties of structural supercapacitors with different salts.

No.	Supercapacitors with Salt	Specific Capacitance ($F.m^{-3}$)	E ($\mu Wh.m^{-3}$)	Flexural Strength (MPa)	Flexural Modulus (GPa)	Hardness (Shore D)
1	Lithium Perchlorate	804	110 ± 1.21	57.4 ± 4.91	28.6 ± 3.62	58.5 ± 4.27
2	Zinc Chloride	12	1.72 ± 0.141	48.8 ± 1.07	26.3 ± 5.82	63.6 ± 6.19
3	Potassium Perchlorate	0.26	0.036 ± 0.004	50.5 ± 1.11	29.8 ± 3.32	55.4 ± 2.87

Table-3**:** Electrical and mechanical properties of structural supercapacitor with a lithium salt variation.

Sample	Specific Capacitance ($F.m^{-3}$)	Energy Density ($\mu Wh.m^{-3}$)	Flexural Strength (MPa)	Flexural Modulus (GPa)	Hardness (Shore D)	Fiber volume fraction V_f (vol. %)
100%R		---- †	191.4± 5.27	87.6 ± 4.97	76.2 ± 4.22	45.8 ± 2.52
5%E	5.1	0.71 ± 0.014	149 ± 6.67	78.1 ± 2.35	73.7 ± 3.76	46.1 ± 1.07
10%E	120	17.4 ± 0.93	114.7±3.68	50.7 ± 8.23	70.4 ± 2.07	48.4 ± 2.12
15%E	564	78.1 ± 1.34	83.4 ± 3.09	37.4 ± 2.3	66 ± 4.02	45.8 ± 1.36
20%E	804	110 ± 1.21	57.4 ± 4.91	28.6 ± 3.62	58.5 ± 4.27	45.4 ± 2.42

† Structural composites with no electrolyte content present.

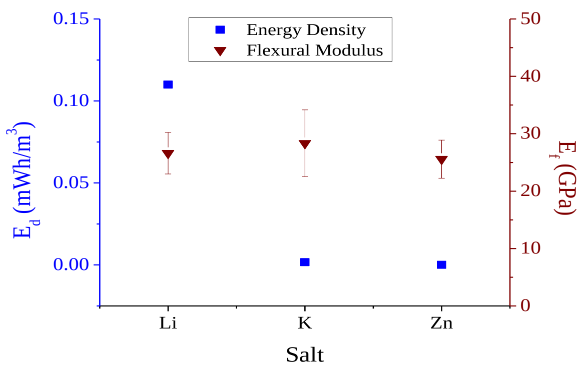

Fig. 4: Energy density and flexural modulus of structural supercapacitors witha salt variation.

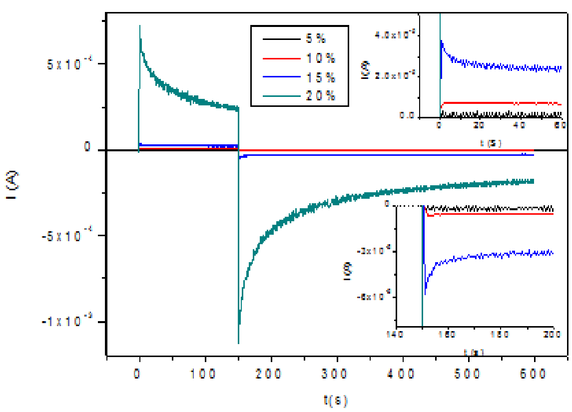

Fig. 5: Charge/discharge plot of structural supercapacitors with varying electrolyte content

Fig 6 shows the optimization of various structural supercapacitor specimens with different compositions of an electrolyte by plotting energy density and flexural modulus on y-axis and supercapacitors with varying electrolyte on x-axis.

By increasing the amount of electrolyte, the energy density of the supercapacitor also increased but with a drawback of decreased flexural modulus. This happened because the increase in electrolyte content decreased the amount of polymer resin, which is responsible for improving the mechanical performance of the supercapacitor, hence a continuous decrease in mechanical properties was observed as the electrolyte content increased from 5% to 20%. Maximum flexural modulus is shown in the Table-3. The 20% electrolyte and 80% resin supercapacitor showed optimum values of energy density and flexural modulus; hence this composition was selected for further studies. Table-3 shows the electrical and mechanical properties of structural supercapacitor with a salt variation.

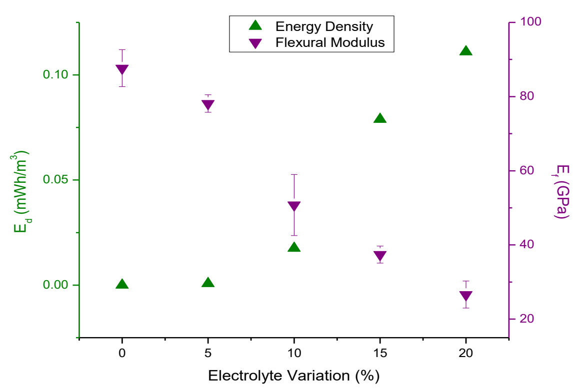

Fig. 6: Energy density and flexural modulus of structural supercapacitors with varying electrolyte content.

Influence of Varying Separator on Mechanical and Electrochemical Properties of Structural Supercapacitor

Glass fiber and filter paper were selected to be tested as an insulating material between carbon fiber electrodes. Influence of varying separator on mechanical and electrochemical properties of supercapacitors was determined. For electrochemical testing of supercapacitors, a charge/discharge test was conducted. It was seen in charge-discharge curve for the filter paper separator, the charge, and discharge area was more than the area for glass fiber based composite. Less area in case of glass fiber based composite was observed because two layers of glass fiber sheets were used to maintain isotropy which resulted in an increase in the separator thickness. With the increase in separator thickness, the specific capacitance was reduced and hence the charge/discharge area and energy density were also reduced. Fig 7 shows the charge/discharge plot of structural supercapacitors with varying separators.

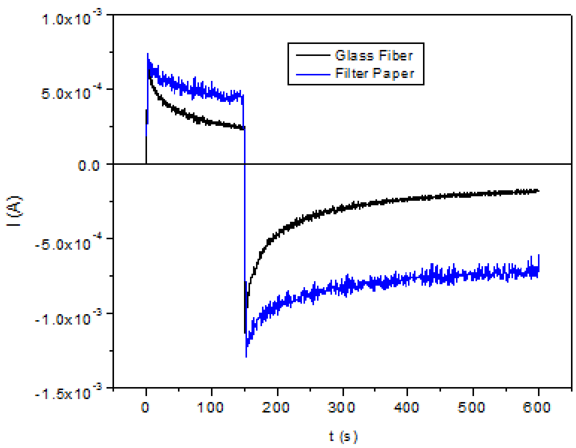

Fig. 7: Charge/discharge plot of structural supercapacitors with varying separator.

In order to examine the influence of varying separator on mechanical properties, the flexural test was performed on each supercapacitor specimen. The stress-strain curve was drawn and on calculating modulus, it was found that due to the presence of two layers of glass fiber, the flexural modulus of supercapacitor with glass fiber, as a separator, increased as compared to filter paper-based structural supercapacitor, because glass fiber gives greater mechanical properties individually. On the other hand, the filter paper is very fragile and has lower strength which shows poor mechanical properties. Fig 8 shows a stress-strain plot of structural supercapacitor using flexural three-point bending tests with varying separator.

Since the electrochemical properties (energy density) of supercapacitor using filter paper were greater than the one using glass fiber as a separator and moreover the mechanical properties (flexural modulus) of the supercapacitor using glass fiber as separator were not very much higher than the one using filter paper as a separator, hence supercapacitor with filter paper as separator was selected for further studies. Table-4 shows the electrical and mechanical properties of structural supercapacitor with different separator.

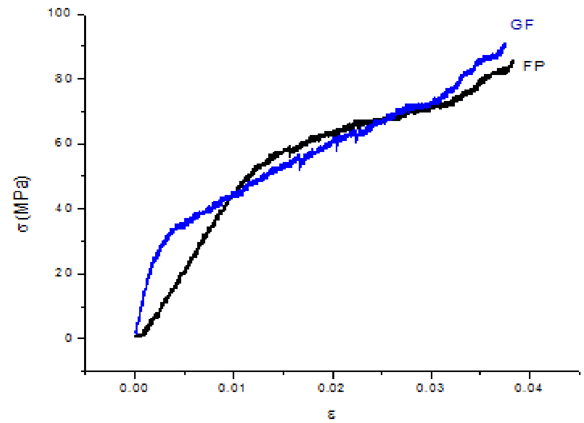

Fig. 8: Stress-strain plot of structural supercapacitors using flexural three-point bending test with varying separator

Multifunctionality of Structural Supercapacitors

Fig 9 correlates the energy density and flexural modulus of supercapacitors of different salts, different salts concentrations, different separators, and different electrodes respectively, as discussed in the previous section. Materials with multifunctional performance in terms of energy density and flexural modulus will lie in the upper right quadrant of the plot. The reference line was also drawn in Fig 9 between the in-plane flexural modulus (38 GPa [11]) of commercial fiber composite and the energy density (1000mWh.m^{-3} [13]) of commercial supercapacitors. All supercapacitors showed a unique combination of energy density and flexural modulus (Fig 9) indicating that these supercapacitors do deliver useful multifunctional performance except supercapacitors having potassium salts electrolyte as they lie below the reference line while all other supercapacitors lie above the reference line.

Table-4: Electrical and mechanical properties of structural supercapacitor with different separators.

Separator	Specific Capacitance (mF.cm^{-3})	Energy density (mWh.m^{-3})	Flexural Strength (MPa)	Flexural Modulus (GPa)	Hardness (Shore D)
Filter paper	**0.80**	**0.11**	**57.4 ± 4.91**	**28.6 ± 3.62**	**58.5 ± 4.27**
Glass fiber	**0.0098**	**0.013**	**62.6 ± 6.85**	**33.6 ± 1.15**	**62 ± 2.56**

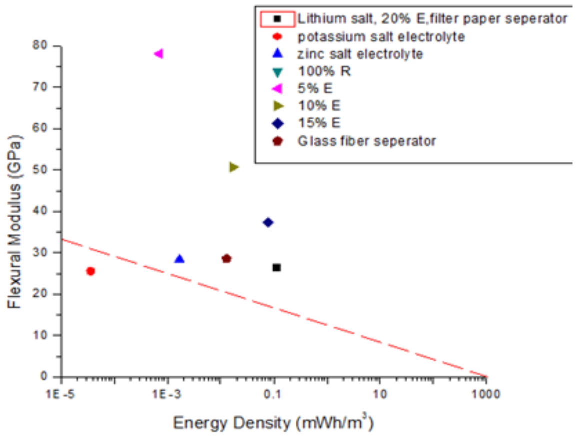

Fig. 9: Multifunctional plot of supercapacitors relating flexural modulus and energy density.

Cost Optimization Chart

The cost optimisation charts of structural supercapacitors were also prepared by plotting the relative cost vs electromechanical performance in accordance with the Ashby diagram [18]. The price of each supercapacitor was determined in $\$.kg^{-1}$. The weight of the capacitor was 0.029 kg. The relative cost, was calculated by dividing the structural supercapacitor cost with the price of steel in $\$.kg^{-1}$. Fig 10 shows the cost of various structural supercapacitor specimens by plotting energy density and flexural modulus on y-axis and supercapacitors with relative cost on x-axis. Fig 10 shows that, by increasing electrolyte content, the energy density and cost increases but mechanical properties reduced eventually. The major expense is of salts so as we increase the amount of salt, the cost of capacitor increases as well as due to increase in the amount of ions which is responsible for conduction also increase, as a result, energy density increases but due to the reduction in resin, mechanical properties decreases. It also shows that supercapacitors having zinc chloride and potassium perchlorate electrolyte are cheaper than lithium salt. To see the multifunctionality, two reference lines were drawn, one of which lies between the plane of relative cost (133[18]) and energy density (1000mWh.m^{-3} [13]) of a commercial supercapacitor. Another reference line was also drawn in between relative cost (133 [18]) and flexural modulus (38GPa [20]) of commercial composites. Supercapacitors with multifunctional performance in terms of energy density, relative cost, and flexural modulus will lie on the upper side of the plot, above the point where the two reference lines meet.

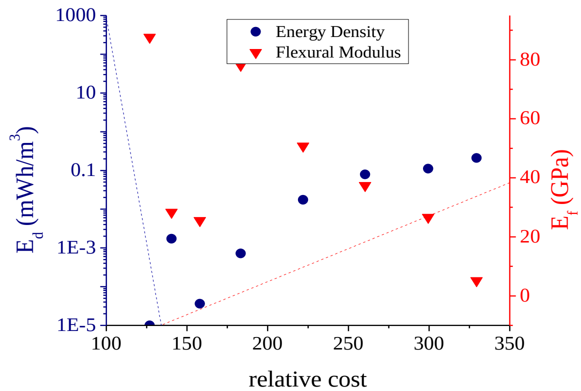

Fig. 10: Cost optimization chart of structural supercapacitor specimens.

Table-5: Relative Cost of different supercapacitors in $\$.kg^{-1}$

Sample	Specific cost $\$.kg^{-1}$	Relative cost[1] $\$.kg^{-1}$
100%R	91.4	126.9
5%E	132.0	183.3
10%E	159.8	222.0
15%E	187.4	260.3
20%E	215.7	299.5
Zinc Chloride	101.1	140.5
Potassium Perchlorate	113.7	157.9

$$\text{Relative cost} = \frac{\text{Price of capacitor}\frac{\$}{kg}}{\text{Price of steel}\frac{\$}{kg}}$$

Price of steel = 720 $\$.ton^{-1}$ = 0.072 $\$.kg^{-1}$ [18]

Conclusion

It can be concluded that the structural supercapacitors are a very economical source of energy which also gives significant structural load-bearing properties. This is the reason why their usage is increasing in automobiles and in other electronic gadgets. In an automobile or electronic gadgets, the major weight is of its battery, so if the body of the machinery is fabricated of such structural supercapacitors, huge weight/volume savings can be attained. The most significant benefit will be the reduction in weight, which, in the case of automobiles, will ultimately lead to less fuel consumption and more mileage. Moreover, these structural supercapacitors can be used in series with another source of energy e.g. conventional batteries, to increase the energy output. In an ultra-battery, a supercapacitor and a battery are combined in one unit thus creating an electric vehicle battery that costs less, lasts longer and is powerful than current plug-in hybrid electric vehicles. Moreover, they are also environmentally friendly; as no redox reactions occur during their operation resulting in longer life cycles. Hence it can be said that the structural supercapacitors are the future energy sources. However, huge efforts are required to increase the specific capacitance of the structural supercapacitors by focusing on improving the multifunctional performance of the individual components. Structural supercapacitors were fabricated by varying electrolytic salt, polymer electrolyte concentrations, separator, and the electrochemical and mechanical performance were evaluated. Structural supercapacitor with simultaneous electrical energy storage of 0.11 $mWh.m^{-3}$ along with a flexural modulus of 28.6 GPa was fabricated and reported here in the current report.

Acknowledgments

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