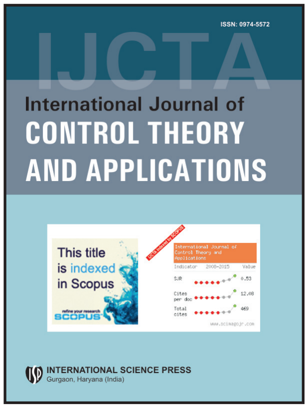

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Agglomeration Reduction in Graphene for Supercapacitors

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Abstract: Here in this work we report how graphene can be prevented from aggregating back to graphite by mixing it with activated carbon at different ratios. The resulting graphene–activated carbon composite of ratio 10:90 exhibited a good specific capacitance of 120 F/g and specific energy density of 6 Wh/kg in KOH electrolyte and 40F/g specific capacitance and specific energy density of 2Wh/kg in KCL electrolyte. The increase in performance can be attributed to the insertion of activated carbon which then acted as a spacer there by increasing specific surface area.

Keywords: Graphene, Supercapacitor, Activated Carbon.

1. INTRODUCTION

Among the challenges faced in this century is unquestionably energy storage. Therefore, it is important that new, environmentally friendly and low-cost energy storage systems be found, in response to the needs of emerging ecological concerns and modern society.

Supercapacitors (also called electric double-layer capacitors or ultracapacitors) are energy storage devices with very high capacity and a low internal resistance, that are able to store and deliver energy at relatively higher rates as compared to batteries due to the mechanism of energy storage which involves a simple charge separation at the interface between the electrode and the electrolyte [1].A supercapacitor consists of two electrodes, an electrolyte, and a separator which isolates the two electrodes electrically. Electrode material is the most important component of a supercapacitor. Some of the benefits of supercapacitors when compared with other energy storage devices are long life, high power, flexible packaging, wide thermal range (–40^{o}C to 70°C), low maintenance and low weight [2].

Different materials such as Carbon materials, Metal oxides and Conducting polymers are currently being used as electrode materials for supercapacitors. Carbon, in its various forms has been used as electrode materials of electrochemical capacitor with the aim of achieving high specific capacitance together with high power density. Although porous carbon materials have high specific surface area, the low conductivity of porous carbon materials is limiting its application in high power density electrochemical capacitors. This further increased the push into nano-structured material where other carbon allotropes were discovered such as Activated carbon, Carbon nanotubes (CNTs) and most recently Graphene [3].

Activated carbon only about 10-20% of the "theoretical" capacitance was observed due to the presence of micro pores that are inaccessible by the electrolyte, wetting deficiencies of electrolytes on electrode surface leading to the inability of a double layer which is necessary for high capacitance to be formed successfully in the pores.

Graphene a two-dimensional single layer of carbon atoms such as graphite has raised a lot of interest since 2004 when it was discovered and keeps making wave in different areas. Due to its unique properties and advantage over other carbon allotropes such as better thermal conductivity, mechanical strength, high specific capacity, surface area, unique heterogeneous electron transfer and charge carrier rates, low cost of production has made it a very lucrative alternative for electrode material[4]. That would bring more technological advancement in the areas like telecommunication devices (cell phones, remote communication, walkie-talkies etc) standby power systems, and electric hybrid vehicles and storage components. Another outstanding characteristic of graphene is its exceedingly high specific surface area up to $2675 m^2/g$. Most importantly is the intrinsic capacitance of graphene that was recently found to be $21\ \mu F/cm^2$. This is the highest value recorded so far in terms of double layer capacitance phenomenon for all carbon-based materials[5].

2. EXPERIMENTAL

The electrode materials were made up of different percentage and ratios to make 1g. This is so that a comparison can be made of the different materials having a definite amount of materials put together. Table 1 below gives a summary of the rations each material took to make up the 1g of electrode material.

Table 1
Percentage ratios of electrode materials

	Material	*Amount/Description*
1.	Carbon Material (Activated Carbon, Graphene)	90% of the total 1g required
2.	Carbon black	2% of 1g
3.	PTFE (binder)	8% of 1g
4.	D.I Water: Isopropanol (solvent)	50%: 50% ratio
5.	Electrolyte: KCL or KOH	3 Molar

Four different samples were prepared in both KCL and KOH, to see the effect of mixing different carbon materials and how the electrolyte affects performances.

Sample 1: AC_ KCL and AC_KOH (Activated Carbon alone 100%).

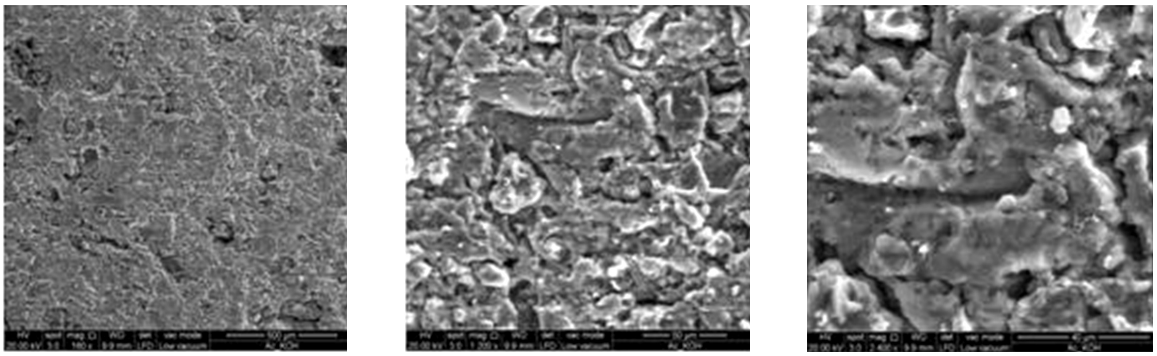
Sample 2: GRN_AC_50_50_KCL and GRN_AC_50_50_KOH (Graphene and Activated Carbon with ratio 50:50).

Sample 3: GRN_AC_90_10_KCL and GRN_AC_90_10_KOH (Graphene and Activated Carbon with ratio 90:10).

Sample 4: GRN_AC_10_90_KCL and GRN_AC_10_90_KOH (Graphene and Activated Carbon with ratio 10:90).

3. RESULTS AND DISCUSSION

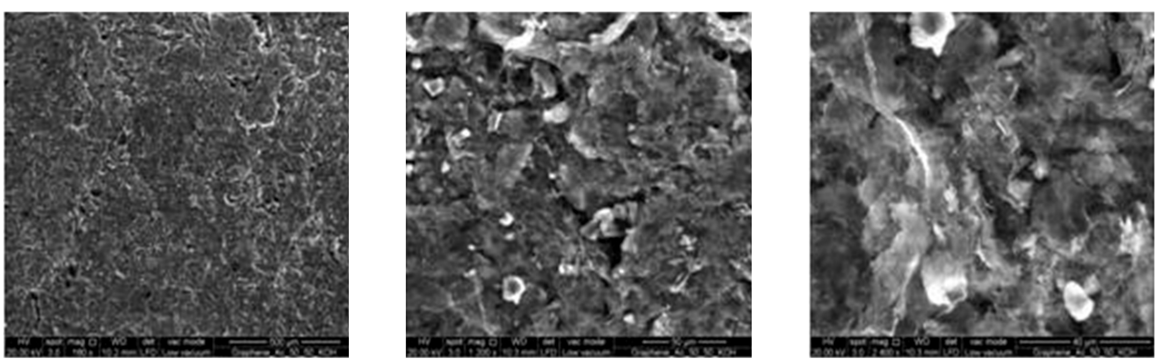
As expected, the result shows the porous structure of activated carbon. Large pore holes can be observed in all three images, the binder effect could also be seen as the particles are mostly joined together around the edges. It was also noted that the carbon black added can be seen at the basal edges of the surface hence the white like structures [6].



(a) (b) (c)

Figure 1: SEM images Sample 1 AC (a) Magnification range of 160 × 500 μm wide. (b) Magnification range of 1200 × 500 μm wide. (c) Magnification range of 2400 × 500 μm wide

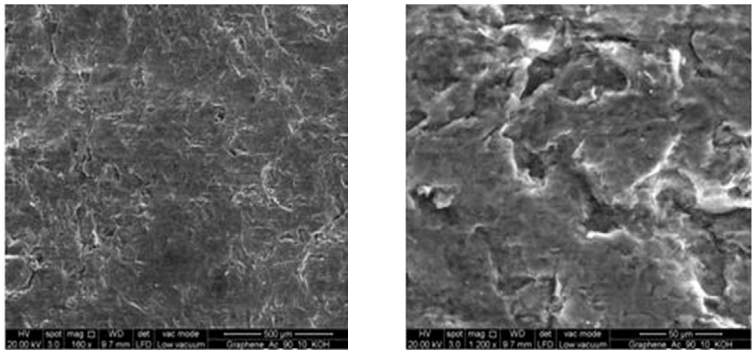
Figure 2 shows the graphene characteristic of slightly crumpled sheets in interconnected vertically with activated carbon. CB is found sparsely on the surface of the electrode sheet and some can be seen to have acted at spacers in between the graphene sheets. In as much as the mixture seems to have both characteristics of AC and graphene, it cannot be said if it would increase or decrease the SSA of the electrode material. In Figure 2(c) the binder effect is seen as a thin line that cuts across the surface of the material joining the sheets together.



(a) (b) (c)

Figure 2: SEM images Sample 2 Graphene/AC (50:50) (a) Magnification range of 160 × 500 μm wide. (b) Magnification range of 1200 × 500 μm wide. (c) Magnification range of 2400 × 500 μm wide

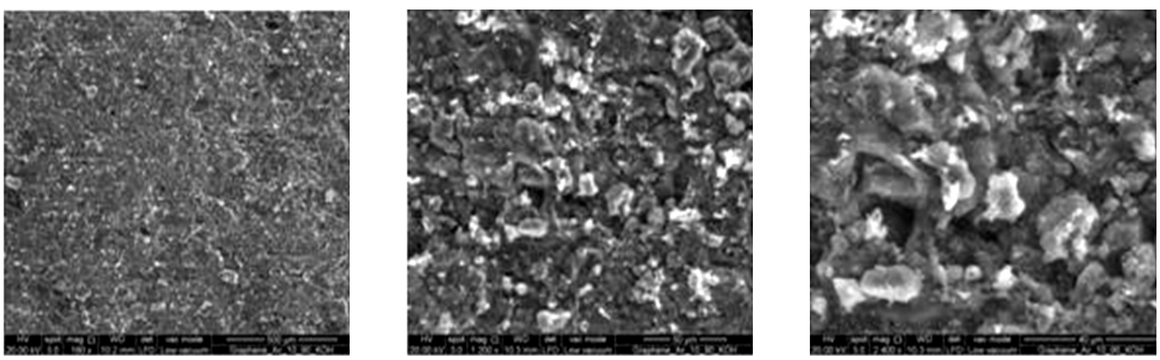
In Figure 3 SEM images of two magnification size were taken showing the surface features of the electrode. This is similar to Figure 2 above except that there are less mesopores and more crumpled interconnected sheets which signify the presence of graphene in larger quantity. Most of the carbon black seem to have been diffused into pores of the electrodes sheet acting as spacers and also increase the conductivity of the electrodes. This also might lead to a larger SSA, specific capacitance and a better energy density. The electrons should be able to easily penetrate into the material not only the surface but also the inner sides compared to the other carbon materials used in the past for capacitors [7].



(a) (b)

Figure 3: SEM images Sample 3Graphene/AC (90:10) (a) Magnification range of 160 × 500 μm wide. (b) Magnification range of 1200 × 500 μm wide

Figure 4 looks quite similar to that of Figure 1 where AC was used 100% as carbon material for the electrode fabrication. CB can be seen on the surface of the sheet, it does not seem to have been absorbed or diffused into the pores of the material but surrounds the edges of the sheets, this would increase the conductivity of the electrodes but not as much as if they were also acting as spacers to spread the sheet further and maybe increase the electron/electrolyte transfer within the sheets. A few graphene layers can be seen at the edges but might not make much difference since they are domineered by the AC materials. Hence sample 4 may not be an appropriate ratio to determine the positive effect of graphene in a mixture with AC material.



(a) (b) (c)

Figure 4: SEM images Sample 4 Graphene/AC (10:90) (a) Magnification range of 160 × 500 μm wide. (b) Magnification range of 1200 × 500 μm wide. (c) Magnification range of 2400 × 500 μm wide

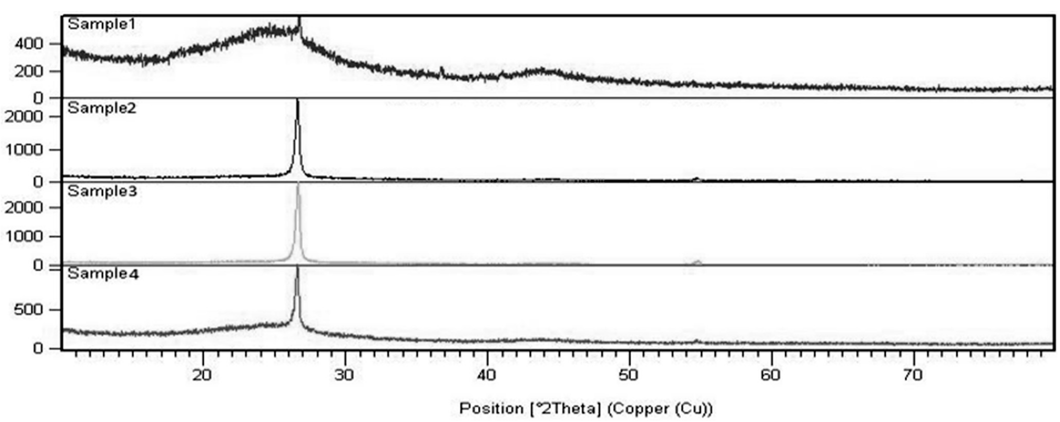


Figure 5: XRD Results

Table 2
Characteristics of XRD patters for the Samples

Sample	*Peak Width Intensity (nm)*	*Position ()*	*Crystal Size (nm)*
Sample 1	0.14	26.70	1.30
Sample 2	0.25	26.59	0.71
Sample 3	0.18	26.62	0.99
Sample 4	0.11	26.56	1.57

Table 3
Specific capacitances and energy density of electrodes

Sample	*Mass of Electrode (g)*	*Electrolyte*	*Specific Capacitance (F/g)*	*Energy Density (Wh/kg)*
1	0.05	KCL	80	2.4
		KOH	90	4.5
2	0.07	KCL	13	0.65
		KOH	23	1.2
3	0.08	KCL	40	2
		KOH	120	6
4	0.05	KCL	11	0.55
		KOH	22	1.10

4. CONCLUSION

In this work, it was established that graphene could not only be used on its own as a material for electrodes in supercapacitors but could be mixed with other carbon materials. By mixing with activated carbon at different ratios the one that yielded the best results was 10:90 of graphene and activated carbon with a specific capacitance of 120 F/g and 6 Wh/kg as energy density this result can be attributed to the increase in specific surface area. Another thing that was proven was the use of electrolyte effects the performance of the supercapacitor. Two electrolytes potassium hydroxide (KOH) and potassium chloride (KOH) were used and it showed KOH to be the better electrolyte as its performance was much higher than KCL this is because KOH is more conductive hence the difference. The specific capacitance and energy density of all the supercapacitors tested using KOH as electrolyte was about almost twice the values gotten from electrodes immersed in KCL electrolyte.

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