

# Comparative Study and Analysis of Asymmetrical Supercapacitors through Modelling Approach

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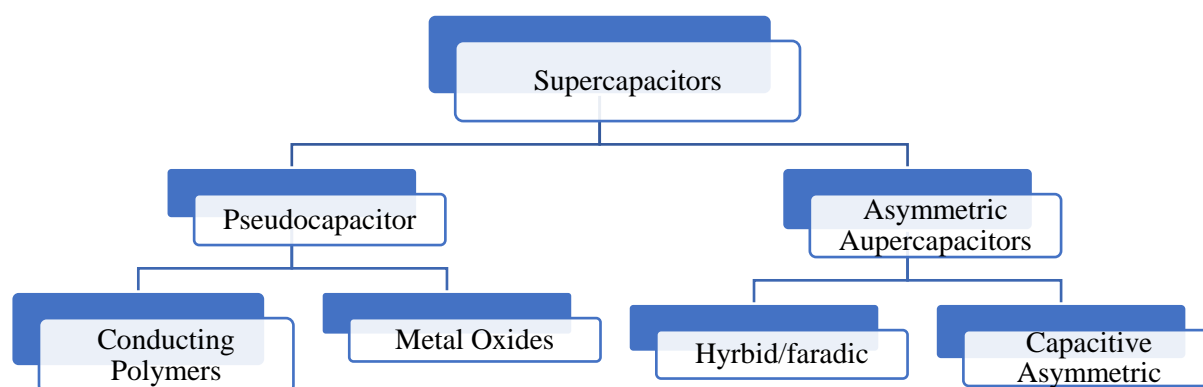
**Abstract:** -It is common with electrical energy that one can find, the times when the supply exceeds the demand, at this condition we use energy storage devices such as flywheels, pumped-hydro, super-capacitor, battery etc which allows energy in a readily recoverable form. Such scenario is quite common in propulsion systems or technology. Asymmetrical supercapacitors are one of the premier upsurging energy storage devices, extensive research is being carried out on these largely useful devices, they take the prestige of the large capacitance which is reintroduce in the vicinity of narrow junction between both porous electrodes and an electrolyte/dielectric. Asymmetrical supercapacitors based on their structure, dielectric material and most importantly on their electrode configuration are widely in research, they are placed with batteries for advantage, because they allow present day high-energy density batteries system to be used effectively, as they loosen up the load of any high discharge currents; are taken up by these asymmetrical supercapacitors. When it comes to any device performance it is very important, to take its modelling and analysis as a priority. In this paper we are presenting how the combination of electrode configuration i.e. vertical and horizontal are affected by electrode parameters in asymmetrical supercapacitors, through statistical modeling.

**Keywords:** Supercapacitor, asymmetrical supercapacitors, modelling, configuration

## 1. Introduction

“Energy” is the one who drives any economy and it sustains every society. The level of affluence of a given country is pertinent related to the per capita energy consumption, which includes food as well as other forms of energy for example electricity. But, keep in mind that its production & usage is the biggest contributor to global warming and many other factors including health issues. The challenge arises when it comes to reduction of the dependence on fossils, conventional/non-renewable energy sources for generation of electricity and also to “store” the generated electrical power. We can’t imagine any sector without electrical power, it makes it very crucial part for social and economic growth. More than half of the world growth in today’s world is due to this “energy”. Switching to more and more to renewable energy sources like wind, solar, tidal, geothermal, biogas, etc. it will serve important role in serving clean energy sources for mankind. Some renewable energy sources are time dependant, intermittent one & some are not. So, in order to use the generated electricity effectively whenever and wherever required we should be able to store them in convenient “energy storage devices”. Li-ion Batteries, flow batteries compressed air storage, mechanical gravity storage, fuel cells, capacitors, supercapacitors etc. are now among top energy storage systems which are been in research which can store energy in either electrical or thermal energy form. Encompassed by the numerous ESD’s, batteries and supercapacitors symbolize the two dominating electrochemical energy-storage technologies [1,2]. Supercapacitor or ultracapacitor, befit in family of energy storage devices with high ratio of out power to total mass and less losses in contrasted to other electrochemical storage devices. They can store energy liquid dielectric called as electrolyte there is no need of or its non-obligatory, to have solid dielectric layer between the two

electrodes. Supercapacitors now adjunct, sometimes oust batteries in a few operations like propulsion system, on the grounds that they almost guardedly bring high power with brisk charging and maximal long cycle life (more than 100 000 cycles) and more. In the boom of this technology, supercapacitors are anticipated to play a pivotal role in the energy-storage sector. The future of supercapacitors looks propitious with opportunities in haulage or area of transport, power tools, and consumer electronics. Research in this range can be anticipated to shoot up with the goal of increasing energy storage [3]. Supercapacitors based on their electrode placement or electrode configuration simply are of two types they are; symmetrical and Asymmetrical Supercapacitors (ASC). The symmetrical supercapacitor which has two same super capacitive electrodes i.e., electrode of same material whereas, asymmetrical supercapacitor has loss of symmetry i.e., they have two super capacitive electrodes but made of two different materials. When it comes to asymmetric supercapacitors, they have two different electrode materials for an example activated carbon at one electrode and manganese dioxide at another or one can have graphene and other can have activated carbon or mixture of graphene and activated carbon. During the charge and discharge, asymmetric super-capacitors have two disparate perspectives or potentials of the two electrodes which lend a hand to enlarge the operating voltage at maximum of the complete device, they also can extend their operating voltage limits beyond the thermodynamic- electrolytic decomposition voltage of occupied electrolyte. Asymmetrical supercapacitors are also classified according to their electrode configuration such as, horizontal and vertical. Both of these have their own advantages and disadvantage based on their material used, material loading, electrolyte type etc. A deservedly designed and analysed asymmetric supercapacitor is the chance of elevate the voltage with parallel increase in energy density for applications. But there is need to study and have deep understanding of various interaction affects in order to increase the specific capacitance.



**Fig.1. Classification (of based on materials and constructions) of supercapacitors.**

When compared to carbon-based materials like hard carbon or activated carbon or any conducting polymers, as mentioned in [4], metal oxides are specifically much advantageous due to their thermal stability and their extra chemical energy storage capability. Material base research on graphene aerogel, activated carbon, graphene nanoplatelets was executed in pursuance to understand the ramification of different carbon-based materials. Graphene-based materials like 0D, 1D, 2D to 3D have proven to be excellent candidates of electrode materials in electrochemical energy storage systems [5-6]. The type of material, development method and staging of electrode materials have evolved the elementary goal of scientific development & research. Also, vertical and horizontal configuration needs to be investigated and studied through modelling approach. Material base research has also been done on graphene aerogel, activated carbon, graphene nanoplatelets was put through in order to master the effects of different carbon-based materials in [5]. There are researches that propose to apply different types of polymers like Poly Pyrrole [7] and also incorporating metal colloids such as gold particles closer near the surface of electrolyte and the porous electrode of supercapacitors have been conducted [7,8]. Manganese dioxide when combined and mixed with various other materials for an ex; The relative dielectric constant and electrical conductivity, rate capability, and excellent cycling reversibility for capacitive charge storage. of electrochemical electrodes with MnO<sub>2</sub> particles mixed with carbon black increases are reported in [9,10,11,12,13]. Doping of various metals in MnO<sub>2</sub>, mixing oxides of Mn and vanadium is also studied, it

improved conductivity, better electrical conductivity than the original form leading to a remarkable enhancement in capacitance [14,15]. Also, increment in the MnO<sub>2</sub> volume fraction boosts the relative dielectric constant, has also been reported [16]. Carbon in dense form resort to its outstanding mechanical, thermal and electrical criterion, its rapport for oxygen at dissimilar temperature scales can be benign utilised in making porous carbons. Greater than 25% of the carbon research in world revolves around activated carbons; newer precursors, to employ high surface area AC from bio-masses such as banana fibre, coconut shells, corn grain, sugar cane bagasse, apricot shell, sunflower seed shell, rice husk etc. an explore as prospective electrode materials for high energy density electrochemical energy storage devices [17,18,19,20] their structure has been studied and experimented which is a benchmark in porous carbon electrode manufacturing and application. Symmetric and asymmetrical supercapacitor with neutral aqueous alkali sulphate have been thoroughly investigated also there is focus on investigating different alkali sulphates, lithium sulphate, sodium sulphate and potassium sulphate, the use of alkali sulphate solutions as electrolytes is a very promising strategy to produce eco-environment friendly carbon supercapacitors, with acceptable temperature performance, excellent cycling life, non-corrosiveness and favor of executing diverse current collectors, making the assembling process easier and their cost is much cheaper than those based on organic electrolytes such as tetraethylammonium tetrafluoroborate [21,22,23]. In this paper is extension of reference [24,25,26]. Research had been done previously to improve specific capacitance of ASCs using fork shaped electrodes. It had proved to improve specific capacitance of ASCs by 300 % [27,28]. ASCs had been previously evaluated for time bound stability in performance based on their configurations [29]. Horizontal/ vertical electrode configuration of ASC is considered with variation in electrode material loading of anode / cathode and percentage of metal oxide in anode as an input parameter for comparative study. Specific capacitance is considered as an output parameter of ASC.

The paper is divided and organised into following sections: Section 2 is oriented towards explanation of all the input-output parameters which are considered for the comparative analysis of model of horizontal/ vertical electrode configuration of ASC; Section 3 has detailed explanation of comparative analysis of statistical models of both vertical and horizontal electrode configuration of ASC; Section 4 presents concluding remarks.

## 2.Objectives

The primary objectives of this research study are to comprehensively investigate and compare the electrochemical performance of asymmetrical supercapacitors (ASCs) with both horizontal and vertical electrode configurations. The study aims to identify the key factors influencing specific capacitance, energy density, and power density in ASCs, focusing on variations in electrode material loading, including activated carbon and manganese dioxide, as well as the percentage of manganese dioxide on the positive electrode. Through a statistical modeling approach, utilizing Full Factorial Design of Experiment (DOE) and Response Surface Methodology (RSM), the research seeks to elucidate the main effects and interaction effects of these parameters on specific capacitance for both configurations. By understanding the intricate relationships and contributions of these factors, the study aims to provide valuable insights into the optimization of ASCs, offering guidance for precise control of electrode material loading to enhance the overall electrochemical performance of these energy storage devices.

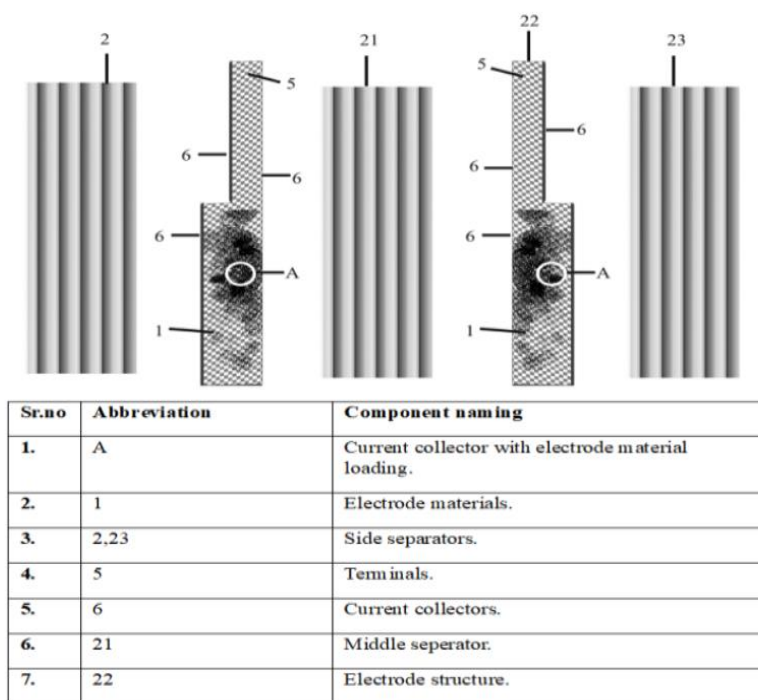
## 3.Parameters Related to Electrodes for Modelling and Analysis

Asymmetrical supercapacitors are just opposite of symmetrical supercapacitors. Asymmetrical supercapacitors have absence of symmetry, i.e. anode and cathode both are of different materials that can be metal oxides, activated carbon, etc. their structure has been studied and experimented which is a benchmark in porous carbon electrode manufacturing and application. In ASCs and even symmetrical supercapacitors the inside as well as outside details are enlightened and studied, each of its component has its own contribution to specific capacitance, power density, impulse current, thermal as well as stress characteristics. ASC has exceptionally commodious no. of device configurations. The pervasive application of supercapacitors has been subjected to limitation because the studies have demonstrated that they have significantly lower energy density than commercially available fuel cells and batteries. Now, with the emerging need to store energy for wearable and portable application in electric propulsion, electronics and power generations etc. research is advancing with the

hope of increasing the application of asymmetrical supercapacitors in order to cope the low density which supercapacitors consist. Any ASC can be identified based on its electrode configuration such as asymmetrical supercapacitor with vertical electrode and asymmetrical supercapacitor with horizontal electrode configuration. Same as the symmetrical supercapacitor the asymmetrical supercapacitor has separator which allows the transfer of ions, and they play the key role to keep electrodes electrically insulated from each other. Many researchers have worked on vertical electrode configurations of anode and cathode but the use of ASC in horizontal electrode configuration makes it new trend energy storage devices in many next-generation electronics portable and flexible structures.

The study on increasing the storage capacity by increasing the area of active material exposed to electrolyte, which in turn determines the operating voltage level has been previously demonstrated. This paper compares the statistical models of horizontal and vertical structured ASC models. This section of paper is reserved for the input parameters corresponding to the two electrodes i.e. anode and cathode of ASC. The input parameters are related to metal oxides and activated carbon, these can vary according to the size and shape of the electrode, molarities and concentration of electrolyte etc. The input parameters are selected based on the model parameters taken from the literature [24,25] and are as follows: a) Loading of activated carbon on negative electrode/cathode b) Loading of electrode material (activated carbon- 50% and manganese dioxide - 50% on positive electrode/ anode) and c) Varying the percentage of manganese dioxide on positive electrode/anode.

The output parameters are energy density, power density, specific capacitance, internal resistance etc. However, specific capacitance being the most important is selected as output parameter. Presented study is about effects of 3 input parameters mentioned, on two types of electrode configuration i.e. on vertical electrode configuration and on the horizontal electrode configuration of asymmetrical supercapacitors. In this paper, models with statistical approach have been used for comparative study of ASC with two type of electrode configuration i.e., vertical as well as horizontal. Fig 2 shows the various parts of the ASC used in horizontal and vertical configuration. Positive electrode consists of activated carbon and manganese dioxide in 1:1 weight ratio. Negative electrode consists of activated carbon as electrode material. Polypropylene sheet is used as separator. Dilute potassium sulphate is used as electrolyte.



**Fig 2 Components of ASC used to get statistical models of horizontal and vertical configurations of electrodes**

#### 4. Comparative Study of horizontal and vertical ASC Configurations

To infer the implicit quality of any energy storage device system its modelling, simulating and testing the models of energy storage system must be performed inclusively and seamlessly. The modelling of energy storage device plays an utmost crucial role in understanding applications of devices. There are various modelling methods and strategies such as mathematical modelling, Theoretical modelling and computational simulation, material-based modelling, statistical modelling, circuit -based modelling etc., researchers make the use of suitable tool for analysis of devices according to available resources. Many researches have made successful attempt in Numerical or mathematical method-based modelling. Theoretical modelling and computational simulation have also been proved to be an effective strategy by researchers. Asymmetrical supercapacitor modelling is an emerging topic and it can lead to advance development in energy storage sector, when it comes to modelling and analysis of this complicated device statistical modelling have been successfully implemented previously in many investigations. Statistical modelling has been success in parameterization in order to understand the accuracy, efficiency and level of details of an asymmetrical supercapacitor. Subsequently on conducting protracted literature reports, it is found that the analysis and modelling of asymmetrical supercapacitor from building outlook is rarely performed and sector related to these asymmetrical supercapacitors are not inspected. Hence, Improved simulation through statistical modelling capabilities will assist in better assessment of which technologies best meet their needs. Numerous other simulations do not justify with renewable energy sources, which in future can undoubtedly be a part of the next generation transmission and distribution systems. Enhanced comprehension of Statistical modelling has worked on other energy storage devices like supercapacitors and batteries and now on asymmetrical supercapacitor.

In modelling of ASC, similar three major input parameters, whereas one output parameter was considered for both Horizontal electrode configuration as well as Vertical electrode configuration, taken from literature [24,25] they are: as per [24] vertical electrode configuration and [25] for the horizontal electrode configuration in the initial stage. There has been consideration in various composition of activated carbon for the negative electrode or cathode and changing percentage of metal oxide, loading of metal oxide and activated carbon on the current collector of positive electrode or anode have been considered. Trials by varying of a single parameter at a time was completed in order to confirm that the parameters considered are affecting the output parameter. Trials are carried out by the variation in loading of electrode material:

2. At first there was variation in loading of (A) activated carbon on the cathode and by managing the loading of activated carbon and manganese dioxide on the current collector of the anode at constant level, in conjunction with the percentage of manganese dioxide on anode to half i.e. 50% at a constant level.
3. Then variation in loading of electrode material (B) i.e. AC and manganese dioxide on the anode and by managing loading of activated carbon on cathode at constant level in conjunction with the percentage of manganese dioxide on anode to half i.e. 50% at a fixed level.
4. In same way, manganese dioxide is varied in percentage on the anode (C) and the total loading of manganese dioxide and AC on the anode is kept at specified fixed level and also the loading of activated carbon on the current collector of the cathode to a constant level.

According to the experimentation on vertical electrode configuration, the results clearly stated that specific capacitance increases relatively right with increases in the percentage of manganese dioxide. Also, all three selected parameters have contribution in affecting specific capacitance with possible presence of some non-linearity. According to the horizontal electrode configuration. Whereas, according to experimentation on horizontal electrode configuration, the results stated that it is found that specific-capacitance is dependent on all three selected parameters as well as there is variation of specific capacitance with material loading on both positive and negative electrode is maximum. Statistical modelling approach is used for studying effect of selected input parameters on specific-capacitance of ASC with horizontally configured electrodes and vertical configured electrodes with positive electrode at the top. The Full factorial Design of experiment (DOE) and (RSM) Response surface methodology method is opted for analysis of the critical parameters. Two level- three

parameters, full factorial DoE and RSM modelling are used for analysis of ASC with horizontally/ vertically configured electrodes. These methods allow user to exactly analyze main and interaction effect in minimum number of trials such as 8 in case of DoE and 20 in case of RSM. This analysis is useful in setting electrode parameter values during the manufacture of these devices. Full factorial DOE and RSM method is a type of statistical modelling a best option for analysis and examining of the critical parameters related to electrodes. The full factorial Design of Experiment (DOE) method for the development of the linear mathematical model, with specific-capacitance as an output parameter. The models represented by equation (1) to equation (4) give main effect and interaction effect between parameters. The Response Surface Methodology (RSM) for developing a quadratic mathematical model to identify the parameter introducing nonlinearity in the device. Now, full factorial DOE and RSM equations with specific capacitance are taken up for comparative study. Table 1 shows Table 1 Effect of various factors on specific capacitance of ASC with horizontal and vertical configuration.

For horizontal electrode configuration:

- DOE equation is:

$$\begin{aligned} \text{Specific Capacitance} \\ = 47.5 - 0.7A + 6.1B - 0.2C - 5.7AB + 3.3AC - 8.1BC - 7.3ABC \quad (1) \end{aligned}$$

- RSM equation is:

$$\begin{aligned} \text{Specific Capacitance} \\ = 21.1 + 3.5A - 4.1B + 0.9C + 3.1A^2 - 3.5B^2 - 0.95C^2 + 0.78AB - 6.9AC \\ - 1.6BC \quad (2) \end{aligned}$$

For vertical electrode configuration:

- DOE equation is:

$$\text{Specific Capacitance} = 26.2 - 1.1A + 7.4B - 1.8C + 2.1AB - 3.9AC - 2BC - 2.8ABC \quad (3)$$

- RSM equation is:

$$\begin{aligned} \text{Specific Capacitance} \\ = 22.3 - 0.4A + 2.4B + 1.5C - 3.5A^2 - 0.45B^2 - C^2 + 4AB + 2AC - 5BC \quad (4) \end{aligned}$$

**Table 1 Effect of various factors on specific capacitance of ASC with horizontal and vertical configuration**

Contributing factors for 2 configurations	Horizontal electrode configuration	Vertical electrode configuration
Main terms	B>A>C	B>C>A
Interaction terms	BC interaction most effective	AB interaction most effective
Non-linear / square terms	A factor contributes to non linear behaviour	A factor contributes to non linear behaviour

#### 4. Conclusions

ASCs are pulse current devices suitable for electric propulsion applications, that employ two electrodes that constitute faradic and non faradic based electrode materials. The anode usually consists of a mixture of highly porous activated carbon and a heavy metal oxide (manganese dioxide in the presented research work). The cathode consists highly porous purely of activated carbon. The research gap that was uncovered with ASCs after an extensive literature review was that ASCs were analysed in both horizontal and vertical configurations but there was no study comparing the performance of the ASCs in these configurations via statistical modelling methods such as DOE and RSM. It has been uncovered through the presented research work that variation of loading of electrode material on the anode (factor 'B') contributes the most towards specific capacitance in both the configurations. This also emphasizes that the factor 'A' (variation of loading of electrode material on the cathode) significantly influence the non-linearity of specific capacitance in both configurations. The factor 'B' also plays a major role in the interaction effect, highlighting the intricate interplay of these factors in



determining the overall electrochemical performance. This emphasizes that research is to be conducted on precise loading of electrode material onto the anode and the cathode to improve specific capacitance of the ASC. For manufacturing of ASCs, loading of material on both cathode and anode should be precisely controlled in order to minimize the variation in capacitance offered by the device manufactured.

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