Investigations related to effect of mass density of metal oxide on specific capacitance of asymmetrical supercapacitor

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Article Info ABSTRACT

Various types of batteries and fuel cells are widely used electrical energy storage devices in most of the applications. Supercapacitors have also become increasingly popular for energy storage in a few applications. Recently the need to develop better energy storage device has resulted in the development of asymmetrical supercapacitors (ASCs). Modeling of this device can be promising to obtain the desired characteristics to meet the need of continuous power and pulse current. For this, both positive and negative electrodes are important components for ASCs. The effect of mass density of metal oxide in positive electrode of this device is hardly researched. Component-based modeling for positive electrode is presented in the paper. Positive electrode parameters are being analyzed with the three most significant parameters affecting specific capacitance. The parameters viz. loading of materials on the current collector, percentage metal oxide, and mass density of metal oxide of the positive electrode are correlated with specific capacitance by using statistical methods to avoid the use of costly electrochemical methods. The major contribution of the presented research work is the identification of – mass density of metal oxide used in positive electrode plays the most significant role in deciding the value of the specific capacitance of this device.

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1. INTRODUCTION

Depletion of fossil fuel reserves and the impact of increasing usage of these fuels are of major concern. However, energy is still needed for the work to be done, and this has constantly led to research on new and improvised technologies for energy storage systems. Supercapacitors and their newer designs are known to have promising results owing to the storage of electrical energy through either double-layer or faradic processes similar to the ones in batteries, or even a combination of both. Supercapacitor banks are integrated with small capacity lead-acid batteries for the development of cost-effective hybrid energy storage systems. The redox processes with transition metal oxides which increase the specific capacitance 10-100 times are classified as supercapacitors [1]. The impact is on the amount of energy stored that can be made available instantaneously. In the categories of the supercapacitor, asymmetrical supercapacitor (ASC) is a device that is capable of providing higher pulse current. Hybrid charge storage mechanisms have also been researched which are a combination of an electrochemical capacitor and an interface of either a battery or fuel cell. Energy storage devices using supercapacitors have been of great interest in recent years. These technologies can replace the traditional batteries and fuel cells in the future. Supercapacitors have been designed using various performance-enhancing electrode materials. Various methods and models have been investigated for modelling of supercapacitors based on discharge behavior [2]. Metal oxide-based Supercapacitors are investigated for improvement of parameters using various manufacturing processes. Research analysis is done with and without binders for high performance testing of Supercapacitors [3]. Efforts are made to construct a low-cost model of supercapacitor using synthesized carbon material [4]. Researchers have experimented with characterization and performance testing for temperature and other important factors of supercapacitors [5]. Due to a need for better energy storage newer designs have been experimented with, leading to the development of ASC [6]. A lot of emphasis is laid on synthesizing new and improved materials for the electrodes of ASC by doping with a combination of vanadium, zinc oxide, manganese dioxide along with mixed-metal hydroxide nanosheets N-doped carbon nanotube array electrodes for high-performance ASCs [7]-[11]. Experimentation is also done on ASC as a pulse current device with its symmetrical as well as hybrid designs [12]. Modern practices for supercapacitors and ASC utilize double layer as well as electrophoretic deposition methods for designing the electrodes [13], [14]. Lattice engineering exfoliation-restacking routes for 2D layered double hydroxide hybridized with 0D polyoxotungstate anions type of cathode are designed for improved hybrid ASC performance [15]. Also, other advanced materials using porous carbon, and interconnected nickel hydroxide nanosheets have improved the performance of ASC [16]-[18].

While designing ASC, asymmetry is an important aspect which is mainly due to different charge storage mechanisms, also due to differences in the amount of charge stored on positive and negative electrodes. Mass and charge balance techniques are the ones that are used for improving the characteristics of ASC [19]. Researchers have experimented with materials that include Nickel alloys for the positive electrode design of ASC [20]. It is observed that the electrode orientation and their placement also have an effect on the parameters which affect the ASC performance [21], [22]. Mathematical modeling is also carried out for hybrid supercapacitors to establish the effect of different parameters on specific capacitance [23]. Various transition metal oxides for positive electrodes are also studied for ASC [24], [25] but component-based modeling is hardly explored. A majority of the literature finds mention regarding negative electrode parameters too. However, positive electrode parameters are less explored which also play a very important role in improving the performance of ASC and this is the research gap. In the presented work, positive electrode parameters are being analyzed with the three most significant parameters viz. amount of loading of material on the positive electrode (A), percentage of metal oxide (B) and the mass density of the metal oxide that is used as electrode material in the positive electrode (C) and they are correlated with specific capacitance by using statistical modelling approaches. An experimental setup is created for studying the effect of parameters of the positive electrode on specific capacitance. The mass density of metal oxide brings difficulties in making stable positive electrode and hence it was decided to study its effect on specific capacitance which is the main innovation of this work. The presented work signifies an impact on the design aspect of ASC with respect to specific capacitance owing to these three considered parameters. Here the work presented is a pouch or stack-type ASC as given in [21] structure by using various metal oxides such as stannic oxide, manganese dioxide and ruthenium oxide. Vulcan XC 72R is used in electrodes and stainlesssteel wire mesh used as a current collector. Potassium sulfate of 0.65 molar strength is used as an electrolyte. The capacitance of the developed module is measured by using discharge characteristics. The results obtained in stack type arrangement can be further extended to the rolled type ASC also.

This paper has five sections: section 2 explains about Taguchi method and the fractional/full factorial design of experiment (DoE) modeling for ASC with specific capacitance as the output parameter; section 3 presents results of the models with discussion related to interpretation of models; section 4 presents concluding remarks.

2. METHOD

2.1. Taguchi model

The Taguchi methodology was employed to investigate and evaluate the performance of ASCs. The design utilized the three variables with three levels of each taken into account consideration for factorial design. Based on extensive experimentation with variation of one parameter at a time, for parameter A, the material loading on the positive electrode, three levels considered are 10 g/sq.cm, 20 g/sq.cm, and 30 g/s q.cm. Similarly, for parameter B, the percentage of metal oxide (weight wise) in the positive electrode, three levels considered are 10%, 50%, and 90%. Lastly, for parameter C, the mass density of metal oxide, three levels considered are 3.36 g/cu.cm, 5.15 g/cu.cm, and 6.97 g/cu.cm. These choices allowed for a comprehensive exploration of the parameter space in the Taguchi methodology. Table 1 shows the coded and actual values at three different levels for Taguchi trials. Table 2 describes the run table for Taguchi trials conducted in the lab.

The model equation for specific capacitance obtained using Minitab software considering the A, B, and C parameters described previously is as (1).

Specific capacitance =
$$
124 - 1.8A - 0.66B + 11.5C
$$
 (1)

Parameter C is most significant and parameter B is least significant in deciding the specific capacitance value.

Table 2. Run table for trials carried out to develop the Taguchi model

2.2. Fractional-factorial model

Using a two-level fractional-factorial DoE model, the performance of the ASC is examined. By employing this strategy, fewer tests compared to the Taguchi methodology are required while capturing the major effects and some inter-parametric interactions. The fractional factorial design utilized the same three variables taken into account. Table 3 shows the coded and actual values for fractional-factorial DoE trials. Table 4 describes the standard run table of fractional-factorial DoE trials conducted.

DoE parameter	Name of the parameter	Min value	Max value
notation		(Actual and coded)	(Actual and coded)
A	Loading of material on positive electrode	10 mg per sq. cm coded as -1	30 mg per sq. cm coded as $+1$
B	Percentage of metal oxide in the positive electrode	10% coded as -1	90% coded as $+1$
	The mass density of the metal oxide	3.36 g per cu. cm coded as -1	6.97 g per cu. cm coded as $+1$

Table 4. Run table for trials carried out to develop fractional-factorial DoE model

The model equation for specific capacitance obtained using Minitab software considering the A, B, and C parameters described previously is as (2).

Specific capacitance =
$$
97.6 - 3.45A - 1.06B + 49.9C
$$
 (2)

As per model (2) , parameter C is the most significant and B is the least significant. This is in line with information extracted from the Taguchi model. Thus, it is clear that mass density of the metal oxide used in positive electrode cannot be ignored. This is one of the major findings of this research work. There is a need to check if this parameter is also playing any role in the interaction effect with other parameters. Hence, full factorial DoE trials were considered as an extension to earlier trials. Full factorial gives information on both the main effect and interaction effect.

2.3. Full factorial DoE model

The same three parameters were considered for full factorial DoE trials. Based on extensive experimentation with one parameter at a time, min and max extreme values were selected for each parameter to define the experimental conditions for two-level, three parameter full factorial DoE. For parameter A, the loading of material on the positive electrode, two extreme values chosen are 10 mg/sq.cm and 30 mg/sq.cm. Similarly, for parameter B, the percentage of metal oxide on the positive electrode, 10% and 90% are selected as the extreme values. Lastly, the extremes of 3.36 g/cc and 6.97 g/cc are selected for parameter C, the mass density of metal oxide. These values are the same as fractional factorial DoE trials. Table 5 shows the standard run table followed in full factorial DoE trials. The model equation for specific capacitance obtained considering the A, B and C parameters using Minitab software is as (3).

 $Specific\ capacitance = 585 - 21.8A - 7B - 49.3C + 0.23AB + 3.8AC + 1.2BC - 0.05ABC$ (3)

Table 5. Run table for trials carried out to develop full-factorial DoE model

Figure 1 shows the surface plots of two level, three parameter full factorial DoE trials. Factors C and A are affecting the specific capacitance as suggested by Taguchi methodology and fractional factorial DoE method. Parameter C is also contributing in interaction effect with other two factors. The critical points where maximum specific capacitance is obtained by considering interaction of all three parameters as per the full factorial DoE model given in (3) were obtained by using the SymPy library in Python. The critical points obtained were as, i) A: 22.7 mg/sq.cm, B: 50%, C: 5.15 gm/cu.cm and ii) A: 20 mg/sq.cm, B: 28.7%, C: 5.15 gm/cu.cm. These are suggested settings for best possible specific capacitance for ASC module.

Figure 1. Surface plots of specific capacitance with variation of parameter A, B, and C (full factorial DOE)

3. RESULTS AND DISCUSSION

The fractional factorial DoE method is implemented to confirm the results given by the Taguchi methodology. The same results concerning the main factors were exhibited by the full factorial DoE method. Two-level, three parameter full factorial DoE model also indicates that there is interaction effect associated with parameter C. Diversity in data for validation is very important. Table 6 shows the randomly selected input parameters used for validating the two-level, three parameter full factorial DoE model. Trials were conducted and specific capacitance was measured experimentally. It is observed that experimental values and full factorial DoE model values match closely.

Metal oxides are mixed with activated carbon in a 1:1 weight ratio. It is found that in order to form a stable positive electrode, mass density of the metal oxide should be on the higher side. This ensures the firm bonding of activated carbon on the metal oxide particles. This has been indicated by the interaction effect to be maximum between parameter A and parameter C. Interaction effect between B and C is also significant as both factors are related to metal oxide in positive electrode.

I able 6. Kun table used for validating full-factorial Doc model								
Input parameters			Specific capacitance in F per sq. cm					
А			By full factorial DoE model Experimental value Error percentage					
0.5	-0.5		0.55	0.53	3.88%			
-1	-0.5		0.3	0.31	5.07%			
0.75	0.25		0.9	0.85	4.91%			
0.75	-0.75		0.82	0.78	4.63%			

Table 6. Run table used for validating full-factorial DoE model

4. CONCLUSION

The fractional-factorial DoE results are the same as those of the Taguchi methodology. It is followed by two-level, three parameter full factorial DoE to get interaction effects. This gives, the positive electrode parameter-based model of ASC with mass density of metal oxide as one of the parameters in it. The points that are the main takeaways from the presented research work are; firstly, conductivity and cost are the main factors in deciding the type of metal oxides to be used in electrodes of electrical energy storage devices like supercapacitors or ASCs. Hence, manganese dioxide is commonly used. But from all three DoE models, it can be seen that the mass density of metal oxide used in positive electrode has a maximum impact on the specific capacitance. There is a need to look beyond manganese dioxide. Secondly, the interaction effect of loading of electrode material and mass density of metal oxide in positive electrode also has a significant role to play in determining the specific capacitance. Hence carbon compatibility with metal oxide to be used should be proper. Thirdly, the percentage of metal oxide on the positive electrode was found to be the least significant parameter in the different DoE models. Lastly, there is a need to look beyond transition metal oxides and replace them with some composite materials with equivalent properties that have better compatibility with the activated carbon used in positive electrode of ASC. Composite materials such as $Co₃O₄$ and Fe₂O₃ with doping, result in metal oxides having higher mass density and lower electrical resistivity that need to be investigated. NiO in its nanostructured form also exhibits similar properties. Such materials could also be conducting polymers, transition metal sulfides and transition metal nitrides. Detailed research work related to it is in progress.

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