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EDLC Capacitance Optimization Using the Taguchi Technique

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Abstract ***- The Taguchi technique was applied in this work to maximize the capacitance of a supercapacitor. The key issue in applying the Taguchi technique is to identify and control the process factors that will lead to optimized capacitance of the supercapacitor. The reported method used in literature is the trial and error method (optimizing one factor at a time). DOE (Design of Experiments) allows multiple parameters to be evaluated with a limited number of experiments. With the use of the Taguchi methodology, in this paper, the number of required experiments was reduced from (3^3) = 27 experiments to only 9 experiments while at the same time producing a more robust product. The main materials used for the fabrication process are activated carbon (AC), which was used as the active material; carbon black (CB) which was used as the conducting agent; and PVDF (Polyvinylidene fluoride) polymer, which was used as binder agent. A total of nine capacitors were fabricated, and the following factors were varied: PVDF percentage, CB percentage and mixing time. This experiment was performed to find the dominant parameter affecting the capacitance. A 1 M (LiClO4) sample in an acetonitrile organic solution was used as an electrolyte, and a 13 mm diameter disc-shaped super-capacitors were fabricated. The cells capacitance was found to range from 24.91 to 51.23 mF with the different process factors. The properties of the fabricated cells were measured using cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS) and charge-discharge measurements. Auto lab (PGSTAT302N) was used to perform the measurements. The results indicated that the binder percentage and mixing time have a significant effect on the capacitance. A final confirmation cell was fabricated using the optimised factors.***

Key Words: **Supercapacitor; Double-Layer Capacitor; Activated Carbon; Taguchi Method; Electrochemical Capacitor.**

I. INTRODUCTION

The Taguchi technique is an experimental design method using "off-line quality control" because it is a method of ensuring good performance in the design stage of products or processes. The Taguchi technique was developed by Genichi Taguchi to improve the quality of manufactured goods, but it can also be applied to biotechnology [1]. With Design of Experiments (DOE) using the Taguchi approach, one can economically achieve product/process design optimization in the manufacturing industry. The objective of using the Taguchi technique is to analyze the outcome of the experiment and then use it as a reference for future experiments. Furthermore, several parameters are set, and samples are made within a limited range, which is altered for each iteration, instead of following the conventional approach of making lots of samples to test each parameter. Once the parameter affecting a process have been determined, the levels at which these parameters should be varied must be evaluated.

Super-capacitors are energy storage devices that have a high energy density, significant power density and a long cycle life [2]. Electrochemical double-layer capacitors (EDLCs) are power sources that store energy within the electrochemical double layer formed at the solid (active material) and the solution (electrolyte) interface [3]. Many different types of electrode materials have been intensively studied for fabricating EDLCs electrodes in recent years. Some of the most used materials include activated carbon [4-6], carbon nano-tubes (CNT) [7-10], carbon aerogels [11] and conducting polymer-based nano composites [12-13].

Activated carbons are recognized as an essential component of the electrode of an electric double-layer capacitor (EDLC) [14-17]. Activated carbon is the electrode material that has been used most frequently for EDLCs due to it many advantages, including low cost, high surface area, availability and established production technologies [18]. The activated carbon grains are mixed with binder, cured (stabilized) and carbonized into activated carbon artifacts before they are connected to the current collector. The form should have a large surface area, the correct pore size distribution and sufficient electric conductivity and mechanical strength. Such properties appear to be governed by the activated carbon even when the binder and forming procedure are carefully selected [19].

At present, because of its stable electrochemical behavior, good cycling performance and low cost, activated carbon has become a mainstream electrode material for commercial super-capacitors [20]. Carbon materials are available with specific surface areas of up to 500–3000 m^2/g [20]. Early electrochemical capacitors were rated at a few volts and had capacitance values measured from fractions of farads up to several farads. The trend today is to construct cells ranging in size from small millifarad-size devices with exceptional pulse power performance up to devices rated at several kilo farads. Some specialized electrochemical cells now in production for traction applications have ratings of more than 100 kF [22]. In this work, the design parameters are the percentage of binder (PVDF), mixing time, and the conducting agent percentage (CB).

II.EXPERIMENTAL

This paper addresses the implementation of Taguchi's technique in the manufacturing process and optimization of a super capacitor. Design of experiments (DOE) can be used to determine the optimal levels for processes or

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design parameters. However, finding these optimal levels is not always an easy task. Industrial experiments do not always go as planned because a nonsystematic approach is often taken by the experimenters. Thus, the following is a step-by-step plan for conducting the experiment, analyzing the results and implementing the solutions [23]:

- Recognition and Formulation.
- Quality Characteristics.
- Selecting Parameters.
- Classifying Factors.
- Determining levels.
- Interactions.
- Orthogonal Array.
- Conducting Phase.
- Analysis Phase.
- Implementation.

In this study, the capacitance (F) of the super capacitor is the desirable quality characteristic. There are other characteristics that can be measured, but the above quality characteristic is considered the most important criterion in this experiment. The chosen control factors and levels that were selected are shown in Table 1.

Table 1. Control Factors and Levels

CONTROL FACTORS /LEVELS	**LEVELS**		
	1	**2**	**3**
PVDF percentage	A1 (2%)	A2 (3%)	A3 (5%)
Mixing Time (Hours)	B1 (1H)	B2 (2H)	B3 (3H)
Carbon Black percentage	C1 (5%)	C2 (10%)	C3 (15%)

The experimental procedure was carried out in a controlled environment in which the experimental conditions remained constant. This ensures that there is no effect on the results from external factors and that the only conditions varied during the experiment are the three control factors, which are binder percentage (PVDF), mixing duration and carbon black (CB) percentage. In particular, room temperature remained constant at 22 °C and the humidity of the environment remained stable during all of the experiments. The same quantities and types of materials were used for all cells. The fabrication process and testing equipment also remained the same.

Taguchi advocates the use of orthogonal array designs to assign the factors chosen for the experiment [24]. Orthogonal Arrays allow one to compute the main effects via a minimum number of experimental trials [25]. Orthogonal Arrays are simple and useful tools for planning industrial experiments. Using an orthogonal array, an experimental plan can easily be constructed by assigning factors to columns of the orthogonal array and then matching the different column symbols with the different factor levels [26]. Because there are 3 factors with 3 levels each, an L_9 Orthogonal Array was chosen. This means that 9 experimental trials with different combinations of the factors should be conducted to study the main effects. In general, the Taguchi design is preferred because it reduces the number of experiments significantly [27]. Table 2 shows the 9 trials, the measured capacitance and the calculated S/N ratio for each cell.

Table 2. Experimental layout

Expt. No.	**Control Factors**			**Capacitance (mF)**	**Mean Sum of Squares**	**S/N Ratio (Larger-the-Better)**
	PVDF %	**Mixing Time (H)**	**Carbon Black %**			
1	A1	B1	C1	33.57	8.87E-04	30.52
2	A1	B2	C2	35.32	8.02E-04	30.96
3	A1	B3	C3	38.36	6.80E-04	31.68
4	A2	B1	C2	41.91	5.69E-04	32.45
5	A2	B2	C3	24.91	1.61E-03	27.93
6	A2	B3	C1	51.23	3.81E-04	34.19
7	A3	B1	C3	43.86	5.20E-04	32.84
8	A3	B2	C1	44.76	4.99E-04	33.02
9	A3	B3	C2	50.75	3.88E-04	34.11

Expt. No.		**PVDF %**			**Mixing Time (H)**			**Carbon Black %**		
	S/N Ratio	**A1**	**A2**	**A3**	**B1**	**B2**	**B3**	**C1**	**C2**	**C3**
1	30.52	30.52			30.52			30.52		
2	30.96	30.96				30.96			30.96	
3	31.68	31.68					31.68			31.68

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4	32.45		32.45		32.45				32.45	
5	27.93		27.93			27.93				27.93
6	34.19		34.19				34.19	34.19		
7	32.84			32.84	32.84					32.84
8	33.02			33.02		33.02		33.02		
9	34.11			34.11			34.11		34.11	
Sum		93.2	94.6	100.0	95.8	91.9	100.0	97.7	97.5	92.4
Factor Effects		31.05	31.52	33.32	31.94	30.64	33.33	32.58	32.51	30.82
OV-Mean	**31.97**	31.97			31.97			31.97		
	31.97	**31.97**	**31.97**	**31.97**	**31.97**	**31.97**	**31.97**	**31.97**	**31.97**	**31.97**

The traditional method of calculating the average effects of factors to determine the desirable factor levels is to look at the simple average of the results. Although the average calculation is simpler, it does not capture the variability of data within the group. A better way to compare the population behavior is to use the mean-squared deviation (MSD) of the results. For the convenience of linearity and to accommodate a wide-range of data, a log transformation of the MSD (S/N) is recommended for analysis of the experimental results [28]. The Signal-to-noise ratio (S/N) is the ratio of the power of the signals to the power of the noise. A high S/N ratio means that there is a high sensitivity with the smallest error in the measurement. In a Taguchi analysis using S/N ratios, a higher value is always desirable, regardless of the quality characteristic [28].

For this study, as the objective is to maximize the capacitance of the super capacitor, the quality characteristics selected for the S/N ratio is the "larger-the-better" (Eq 1) [32].

Signal-to-noise (S/N) ratio = $-10 \log_{10}((1/y_1^2 + 1/y_2^2 + \ldots\ldots + 1/y_n^2)/n)$ (1)

Where n= number of values at each trial condition and y_i= each observed value.

III.RESULTS & DISCUSSION

In Taguchi's parameter design, the basic objective is to identify the conditions that optimize the process/ product performance. In arriving at this optimal set of conditions, Taguchi advocates the use of the signal-to-noise ratio (S/N ratio). The goal is to maximize the performance of a system or product by minimizing the effect of noise [23]. The S/N ratio is treated as a response (output) of the experiment, which is a measure of variation when uncontrolled noise factors are present in the system [23].To maximize the S/N ratio, (which in effect means, to reduce the quality loss caused by variability about the nominal value), we need to do two things:

- Select values for the control factors that minimize variability. In this way, we minimize the effect of the noise factors, that is, we aim to make the process 'robust'.
- Select values for the control factors that change the mean value of the quality characteristic to correspond to the nominal value.

The analysis will therefore be in two parts:

1.Identify which control factors have the greatest effect on variability. Select values for these control factors accordingly.

2.Identify which control factor has the greatest effect on the mean, other than those already set to minimize the variability. Select a value for this control factor to bring the predicted value of the quality characteristic as near as possible to the nominal value.

Table 3 and 4 illustrate the average S/N ratio values and the average means values, respectively, for the capacitance at three levels settings of each factor and the effect of each main effect on the S/N ratio.

Table 3. Response Table for the Capacitance S/N ratio

	A	B	C
Level 1	3.102	3.181	3.229
Level 2	3.068	3.012	**3.231**
Level 3	**3.328**	3.**316**	3.028

Table 4. Response Table for the Capacitance Means

	A	B	C
Level 1	35.750	39.780	**43.186**
Level 2	39.350	34.996	42.660
Level 3	**46.456**	**46.780**	35.710

Figure 1 illustrates the main effects plot of the control factors for the experiment (using the values from Table 3).

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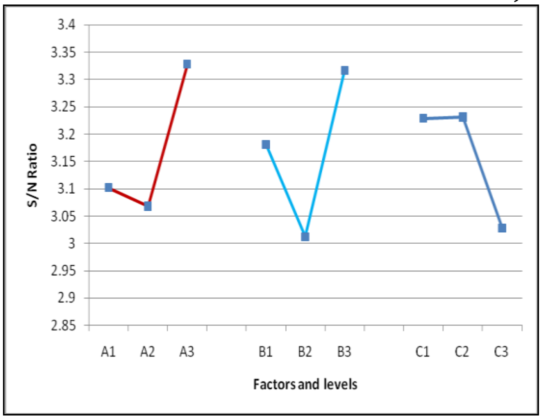

Figure 1. Response Graph for the S/N ratio (Capacitance)

Figure 1 and Table 3 show that the most significant factor for Capacitance is control factor A (PVDF %). The effect of factor B (Mixing time) is also significant, but factor C seems to be less significant. Because the objective of this work is to optimize the capacitance, we need to maximize the S/N ratio to minimize the variability. Thus, both factors A and B need to be set to level 3.

Analyzing the results from the response Table and graph reveal that there is a small difference between the results already mentioned for the capacitance optimization. In particular, Table 5 and Figure 3 show that the most significant factors for increasing the capacitance are control factors B (Mixing time) and A (PVDF %). The effect of factor C is less significant. The factors need to be set to A3, B3 and C2 to obtain the maximum capacitance measurement.

Control Factor	**Level 1**	**Level 2**	**Level 3**	**Opt Level**
PVDF %	A1 2%	A2 3%	A3 5%	**A3 5%**
Mixing Time (H)	B1 1H	B2 2H	B3 3H	**B3 3H**
Carbon Black %	C1 5%	C2 10%	C3 15	**C2 10%**

Finally, it can be observed that the most important factors for the capacitance optimization are factors A and B. The experiment enabled the behavior of the system to be understood by the engineering team in a short period of time and resulted in significantly improved performance (with the opportunity to design further experiments for possible greater improvements) [31]. At the end of the experiment, a final confirmation experiment was performed, and three super capacitor cells were manufacture using the optimized factors: pvdf percentage (5%), mixing time (3H) and carbon black percentage (5%). The capacitances of those cells were 54.57, 53.89 and 55.65 mF. The average capacitance of the cells is 54.70 mF.

IV. CONCLUSIONS & FURTHER EXPERIMENTATION

The work aimed to apply Taguchi's Experimental Design for the optimisation of super capacitor capacitance. Taguchi's method of experimental design has been proven to be a powerful tool for the optimization of product designs or processes because it can provide an experimenter with the maximum amount of information possible from the least number of experiments [29]. It was clearly demonstrated that the Taguchi DOE methodology is applicable in super capacitor optimisation studies. To study the effect of variables in the minimum number of trials, the Taguchi approach to experimental design was adopted [30]. The project illustrated the techniques that can be used to achieve the best outcome at the minimum cost. With regard to the EDLC under study, the results indicated that binder percentage, mixing time and conducting agent percentage are some of the significant factors that can provide an optimised EDLC if adjusted properly. The optimisation of various processes (such as the mixing process, coating process and drying process) could be the next step in terms of further experimentation, followed by an investigation of different raw material (AC, BC and binders) base mixes.

The analysis of the results obtained using Taguchi's method showed that this method is one of the fastest and most reliable, effective, beneficial and economical ways to optimize an experimental design and find the appropriate conditions to obtain an extremely high-quality product. This method can be applied to other important design characteristics and attribute quality characteristics, which will also yield interesting results.

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