Research Article

Chicken Eggshell-Based Capacitors: Fabrication and Evaluation of Their Performances for Engineering Application

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Received: 27/Oct/2024; **Accepted**: 28/Nov/2024; **Published**: 31/Dec/2024

Abstract — Chicken eggshells are usually generated in very quantities but majorly under-utilised. This situation warrants discharging them indiscriminately into the environments. Consequently, this disposal technique pollutes the environment and also causes breeding of disease-carrying organisms, thus posing severe adverse effects on public health. In this work, chicken eggshells were gathered and processed into powdery form at three different ages (storage times) such as 5, 15, and 30 days. The chicken eggshell powder (CEP) in each case was used as dielectric material to fabricate capacitor. The dispersing medium used in the fabrication process was slurry prepared from dry cassava starch (DCS). Five capacitor samples were developed for each considered age. The CEP, DCS, and fabricated capacitor samples were assessed for their potentials. The Carr's index of the CEP was found to be approximately 9.00 % whereas that of the DCS was about 11.41 %. Over the temperature range of 20 $^{\circ}$ C to 70 ^oC, the capacitance of the capacitor samples decreased from 8.93 to 4.62, 7.66 to 2.15, and 5.59 to 1.84 (all in nF) for the eggshells processed at the age of 5, 15, and 30 days respectively. The temperature coefficients of capacitance for the same trend in age variation were - 0.97, - 1.44, and -1.44 (%/°C) based on JIS standard and also – 0.90, - 1.39, and – 1.34 (%/°C) based on EIA protocols. The overall relative permittivity decreased from 9137 to 1883 as temperature of the samples increased over the considered range. Statistically, the relative permittivity values between the CEP of 15 days and that of 30 days only were revealed to be insignificantly different. The capacitor samples exhibited good performance abilities when compared experimentally with conventional ceramic capacitors.

Keywords — Capacitance, Cassava effluent, Flowability, Recycling, Relative permittivity, Waste, Storage times

1. Introduction

Agriculture is one of the sectors from which large quantities of wastes are generated yearly. Due to the persistence of ineffective solid waste management systems in developing and less-developed countries, there have been serious concerns on how to properly handle the wastes in order to protect the environments and also ensure healthy living [1]. Interestingly, recycling has become a safe and beneficial option for solving the attendant disposal problems. This, indeed, offers an opportunity for the bioeconomy society [2]. A typical example of agro-waste that is characterised by recyclability, sustainability and potentiality is chicken eggshell. Chickens (*Gallus domesticus*) and man have coexisted for several millennia. Keeping of chickens by humans is primarily as a source of food, consuming both their meat and eggs. The adoption of technological innovations in agriculture has made commercial poultry farming to become a fast-yielding business in recent times. For instance, the practice of animal breeding facilitates the possibility of producing commercial chickens (known as layers) that can lay several eggs within few days unlike how local (indigenous) breed can do [3]. Chicken eggs can be of optimum size as well [4].

Laca et al. [2] described chicken eggshell as a natural porous bioceramic resulting from the sequential deposition of different layers around the albumen in final sections of the hen oviduct. Chicken eggshell represents approximately 10 - 11 % of the whole egg weight [5]. It is a potential raw material for certain applications which mostly aim at development of entirely new products. Chicken eggshells are used for making bone graft substitute/hydroxyapatite [6], [7], [8], [9], and wastewater treatment [10], [11], and as source of calcium [12], [13], [14]. Other studies have revealed that chicken eggshell powder is useful in the development of composite panels [15], [16], [17], [18], and preparation of electrode [19], [20], [21], as well as manufacturing of temperature sensing/monitoring/control devices [22]. The growth in Africa's egg industry is estimated to be 3.9 % annually, having Nigeria as the biggest market in sub-Saharan Africa with an ongoing growth of 3 % to 5 % predicted for the period of $2017 - 2027$ [23]. As eggshells remain underutilized, a large amount is disposed of by burning and this practice is severely harmful to the ecosystem thereby becoming a worrisome issue as it contributes to the indices of challenges confronting development of the economy. As such, it is necessary to explore a convenient way of managing waste chicken eggshells for beneficial purposes, especially,

now that the conventional materials for fabrication of capacitors have become expensive.

The aim of this study, therefore, is to fabricate capacitor form waste chicken eggshells of different ages and assess the performance of the developed products in comparison with conventional ceramic capacitors. Importance properties like capacitance, dielectric permittivity, and temperature coefficient of capacitance will be assessed for the eggshellbased capacitors. To the best of the authors' knowledge, no such scientific information is available in the literature, making this paper to be the first in reporting the development. Capacitors have all sorts of critical applications in circuits and they play important roles in electronic systems [24]. It is hoped that the findings from this study would serve as a guide on how to solve the disposal problems associated with eggshells while providing useful insights to practitioners, researchers, manufactures of electronic devices, and designers of electronic systems on ways to enhance growth and development of electrical/electronics industry in particular, and the nation's economy at large.

2. Related Work

In recent times, efforts have been made by some researchers on the possibility to produce capacitors from certain waste materials. Ma et al. [25] applied a two-step approach to make super capacitors from waste wood through pyrolysis and optimization of chemical activation conditions. They found that carbon monoliths activated by potassium hydroxide exhibited an optimal specific capacitance of 95 Fg^{-1} , due to their high specific surface area (424 m2g^{-1}) and rich micropore volume $(0.13 \text{ cm}^3 \text{g}^{-1})$. It was observed that the total capacitance $(4.7 \text{ F}$ for 1 cm^2 of one electrode) was much higher than that of common pellet electrodes (about 0.5 F) although the specific capacitance was not highly competitive. At last, technical-economic assessment confirmed the potential for energy saving from an industrial perspective, and a cost breakdown of \$ 4.6 per kg was expected in electrode assembly process. Globally, this strategy has the potential to be used for many other waste woods and holds much promise for the advancement of sustainable energy industries.

Bhat et al. [26] reported on the general concepts of new developments in production of high-value activated porous carbon from various types of wastes for use in supercapacitors. They noted that the composition of diverse wastes (including E-waste, tea, palm, plastic, agricultural, and animal wastes) has made waste materials viable candidates for the starting substance of activated carbon for supercapacitor performance. Apart from discussing the advantages of several types of waste materials utilized as energy storage, they addressed the procedures for preparing activated carbon (such as pretreatment and activation) as well. Finally, they highlighted the economic element of supercapacitor generation from trash, as well as future potential for supercapacitor application. In conclusion, they proposed environment friendly use of waste materials by using activated carbon for energy storage via supercapacitors

Jekal et al. [27] adopted recycling to successfully fabricate all-solid-state asymmetric supercapacitor (FASC) device from heated tobacco waste (HTW). This involved carbonization of cellulose acetate tubes and tobacco leaves, after which they were mixed with metal oxides $(MnO₂$ and $Fe₃O₄$) to obtain highly active materials for supercapacitors. Moreover, they dissolved poly (lactic acid) (PLA) filters in an organic solvent and mixed with the as-prepared active materials using a simple paste mixing method. Additionally, flexible $MnO₂$ and Fe₃O₄-mixed HTW- C/PLA electrodes $(C-MnO₂/PLA$ and $C-Fe₃O₄/PLA$ were successfully fabricated using the drop-casting method. It was noticed that the as-synthesized flexible C-MnO₂/PLA and C-Fe₃O₄/PLA electrodes exhibited excellent electrical conductivity of 378 and 660 μ S cm⁻¹, and high specific capacitance of 34.8 and 47.9 mF cm⁻² at 1 mA cm⁻², respectively. A practical FASC device $(C-MnO_2/PLA/C-Fe_3O_4/PLA)$ assembled by employing the $C-MnO₂/PLA$ as the positive electrode and C - $Fe₃O₄/PLA$ as the negative electrode showed a remarkable capacitance of 5.80 mF cm⁻² at 1 mA cm⁻². In addition, the FASC device manifested stable electrochemical performance under harsh bending conditions, verifying the superb flexibility and sustainability of the device.

Sandhiya et al. [28] synthesized N-doped porous activated carbon derived from poultry waste (PW) for the fabrication of a flexible supercapacitor. They found that the specific capacitance proliferated 7.2 times after chemical activation with potassium hydroxide (KOH). After activation of PW material, the energy density of symmetric supercapacitor snowballed significantly from 16 to 23 Wh per kg. In addition, the durability of the supercapacitor enhanced from 83 to 99 % after activation. The enhancement in the specific capacitance and energy density was noted after the activation process due to the high surface area of PW-derived carbon by activation with KOH. The enhancement in cycle life has been observed due to the high degree of graphitization. Moreover, an all-solid-state flexible supercapacitor (ASSFSC) fabricated using N-doped activated carbon showed an enormous energy density of 21.5 Wh per kg. The results demonstrated that the supercapacitor fabricated using activated PW-derived carbon could be commercialized easily.

Sohail et al. [29] reported on ceramic wastes based composites for capacitors applications. They found the wastes to be 50 % efficient in removing methyl orange from water in a specific time. This suggested that the prepared materials can be used in energy harvesting appliances (i.e., capacitors) while the ceramic waste can be applied for purifying polluted water coming out from industrial as well as municipal sewerages. Nanaji et al [30] explored bio-waste plant stemderived activated porous carbon as a cathode for Li-ion hybrid capacitor (LIHC) application to achieve a high power and energy-based system. It was noticed that a specific surface area of 1826 m^2g^{-1} enhanced degree of crystallinity and graphitization results for porous carbon from activation by from activation by potassium hydroxide. When employed as supercapacitor material, the device exhibited good rate capability, energy, and power attributes with a specific capacitance of 116 F g^{-1} (1 A g^{-1}). Simultaneously, when tested for LIHC application the formulated device showed good capacity retention for 2500 cycles with a high energy density of 125 Wh per kg at a power density of 69 W kg⁻¹.

Mikolajek et al. [31] presented the preparation of fully inkjet printed capacitors containing ceramic/polymer composites as the dielectric material. This involved development of eramic/polymer composite inks, which allowed a fast onestep fabrication of the composite thick films. $Ba_0Sr_0ATiO_3$ (BST) was used as the ceramic component and poly (methyl methacrylate) (PMMA) as the polymer. The use of such composites allowed printing on flexible substratesand resulted in improved values for the permittivity compared to pure polymers. Three composite inks with varying ratios of BST to PMMA were used for the fabrication of composite thick films consisting of 33, 50 and 66 vol % BST, respectively. All inks led to homogeneous structures with precise transitions between the different layers in the capacitors. It was found that the printed capacitors exhibited dielectric constants of 20 up to 55 at 1 kHz.

Challagulla et al. [32] demonstrated a facile, cost-effective, green and sustainable fabrication of supercapacitor device by using high surface area (2350 m^2g^{-1}) activated carbon fibres as supercapacitor electrode. The electrochemical behavior of the supercapacitor electrodes with different neutral electrolytes such as lithium chloride (LiCl), potassium chloride (KCl), and sodium chloride (NaCl) were carefully investigated and compared with natural seawater as economic and sustainable electrolyte for the first time. The maximum specific capacitance of carbon fibres electrode in different electrolytes were found to be around 101 Fg $^{-1}$ in LiCl, 134 Fg^{-1} in KCl, 159 Fg^{-1} in NaCl and 172 Fg^{-1} in natural seawater at a current density of 1 Ag⁻¹. Surprisingly, seawater based supercapacitor exhibited a very good durability upon 10,000 charge-discharge cycles with 99 % of capacitance retention and also 99 % of coulombic efficiency. For practical validity, the integrated solar cell based supercapacitor pouch cells were investigated in which the seawater was explored as an eco-friendly, cost-effective and alternative aqueous electrolyte that may replace the aqueous based electrolytes for the fabrication of economic and green supercapacitor device.

The study by Vijayakumar et al. [33] delineated the fabrication and electrochemical performances of activated porous carbon fibers used as high performance supercapacitor electrodes with commercial level mass loading $(150 \pm 10 \,\mu m)$ thickness, 10 ± 1 mgcm⁻²). The fabricated supercapacitor electrodes showed combination of high gravimetric and volumetric capacitances in three different electrolytes 6M KOH (0 - 1 V), $0.5M$ Na₂SO₄ (0 - 1.8 V) and 1M TEABF4/AN $(0 - 2.7 V)$ due to the thick electrodes (high active mass loading). Particularly, the electrodes in organic electrolyte TEABF4 at 2.7 V exhibited excellent gravimetric and volumetric capacitances of 112 Fg^{-1} and 74 Fcm^{-3} at 1 Ag⁻¹, respectively with long life cycle (87 % capacitance retention after 10,000 cycles).

Dědek [34] successfully demonstrated how to obtain a highperformance supercapacitor from plastic waste. This study

focussed on a two-step process that involved pyrolysis followed by chemical activation, which effectively transformed common plastic waste into activated carbons (ACs), making them suitable for electrode materials in supercapacitors. In addition to well established parameters such as specific surface area and micropore volume, the research highlighted the importance of other critical factors, including the glass transition temperature of the polymer, compatibility between the polymer and activating agent, the ratio of activating agent (K_2CO_3) to ACs, and the stability of ACs in water dispersion. ACs with a competitive electrochemical performance were obtained. Specifically, the ACs exhibited a specific capacitance of 220 Fg^{-1} (at a current density of 1 Ag^{-1} , energy and power densities of 61.1 Wh per kg and 36.9 kW per kg, respectively, and excellent cycling stability (95 % retention after 30,000 cycles). The findings provided a pathway towards transforming plastic waste into valuable electrode materials for supercapacitors.

Zhang et al. [35] transformed polystyrene into threedimensional (3D) network structure porous carbon (PC) via the Friedel–Crafts reaction. The constructed carbonyl (– CO–) cross-linking bridges between the linear polystyrenes provided the resulting hierarchical porous polystyrene with a high cross-linking density and amounts of oxygen atoms to achieve the carbonizability of cross-linking polystyrene framework. Moreover, silica particles created more porous structure for carbon material. The prepared PC showed large specific surface area and 3D porous structure and exhibited good capacitance and electrochemical stability as electrode materials for supercapacitor.

Lian et al. [36] reported a successful design and synthesis method for developing a graphene/mesoporous carbon (G@PE40-MC700) electrode materials from upcycled waste polyethylene (PE) plastic combined with graphene oxide (GO) and flame retardant by low-temperature carbonization at 700 °C. The G@PE40-MC700 exhibited a high surface area $(1175 \text{ m}^2 \text{g}^{-1})$ and a considerable amount of mesopores $(2.30 \text{ cm}^3 \text{g}^{-1})$, thus improved electrochemical performance in both symmetric and hybrid supercapacitors with wide voltage windows. The hybrid supercapacitor assembled from $G@PE40-MC700$ as anode and $LiMn₂O₄$ as cathode operating at 2.0 V in 0.5 M $Li₂SO₄$ was investigated. The hybrid supercapacitor delivered an energy density of 47.8 Wh per kg at a power density of 250 W kg^{-1} , as well as high cycling stability of 83.8 % after 5000 cycles.

3. Methodology

3.1 Materials

The materials used in this study included chicken eggshells, cassava effluent, cotton wool, tap water, and conventional ceramic capacitors. The chicken eggs were procured from a local poultry farm (Niger Delta Basin, Shungai). Care was taken to ensure that the eggs used were laid by a particular chicken and its dietary was not varied.

3.2 Processing of the chicken eggshells

The chicken eggs were first stored at room temperature and then, at various ages (storage times) such as 5 days, 15 days

and 30 days, before they were de-shelled. The de-shelling was done without heat treatment and the shells were washed thoroughly in running tap water before they were sun-dried. The dried eggshells were then stored separately in waterproof and labelled/coded for ease of identification. After that, each category of the eggshells was crushed into powdery form using Agate mortar and pestle. The crushed material was sieved and then calcined at $1000\degree$ C for 6 hours using ASCO Muffle furnace. On cooling to about 35° C, the calcined material was pulverized at 500 rpm for 5 hours by means of High-energy ball miller (Emax, manufactured by RETSCH, GmbH). According to the manufacturers, this ball miller can reduce particle size of a material feed from about 5 mm to as fine as less than 80 nm. The cassava effluent was used to obtain starch for preparation of the binder needed in this study. Figure 1 shows some of the raw materials and their processing stages.

3.3 Analysis of the chicken eggshell powder (CEP)

Following the standard procedure outlined in ASTM D 6393 [37], the Carr's index of each CEP material was determined for determination of flowability. The determination was done in triplicates. In this study, 100 g of each CEP material was weighed at a time by means of a digital balance (Model CS 2000) and transferred to a 100 cm³ glass measuring cylinder. The bulk volume occupied by the material at that instant was noted before the cylinder was tapped 50 times using a tapping rod. After the tapping process, the tapped volume of the material was noted. Also, bulk density and tapped density of the material were calculated as the ratio of mass to the respective volumes. Based on the values of bulk density, and tapped density obtained, the corresponding Carr's index of the material was computed thus

$$
CI = \left[1 - \left(\frac{\rho_b}{\rho_t}\right)\right] 100\,\,\%
$$
 (1)

where $CI = Carr's index$, $\rho_h = bulk density$, and $\rho_t = tapped$ density.

3.4 Fabrication of CEP-based Capacitor samples

Each CEP material was mixed with binder/slurry in order to make it easy for processing. The mixing ratio of the CEP to the binder was maintained at 3:1 by weight. The binder was prepared from dry cassava starch (DCS) derived from the cassava effluent via decantation and drying as detailed by Robert et al [38], Like in the case of the CEP, the flowability of the DCS was assessed. The DCS was dispersed in water to obtain a 15 % $\frac{w}{v}$ solution, which was heated until it became gummy. The slurry (gummy liquid obtained) was poured onto conveyor belt inside a drying oven, resulting in the dry tape. This was then cut into pieces called sheets. The electrodes were fixed to the sheets after which the sheets pressure was applied to create a monolithic structure called a bar. With the aid of the saw, the was cut into separate capacitors known to be in green state before being fired in the kiln with slow moving conveyor belts. Figure 2 summarizes the fabrication processes adopted in this study. For each CEP material, five identical capacitor samples were fabricated and used in assessment of the basic characteristics and performance. The thickness of each fabricated capacitor was 0.16 cm. All the

capacitor samples were coated to make their surfaces moisture-resistant.

Figure 1. Preparation processes of the CEP and DCS

Figure 2. Flow diagram of the CEP-based capacitor fabrication stages

3.5 Testing of the Capacitor samples

For performance evaluation, **t**he capacitance of each sample was checked at 20 °C by means of an LCR meter (Model No. 9183, Lutron) and noted. Conventional ceramic capacitors having equivalent/very similar capacitance values were then selected. This was necessary to ensure proper comparison of the performances of the fabricated capacitors with the commonly-used ceramic capacitors. The performance was considered to be in terms of their signal smoothening ability based on the working of a half-wave rectification circuit. This circuit consisted of an alternating voltage (AC) source, a power diode (1N5401, Silicon), load resistor (10 M Ω), and capacitor. Figure 3 shows the circuit diagram and the layout diagram of the components on a breadboard.

The audio signal generator (Model 2224-1) was employed to supply voltage at 50 Hz to the anode of the diode and a terminal of the capacitor linking one end of the load resistor. Figure 4 shows some of the CEP-based capacitors conventional ceramic capacitors, and test setup features. During the assessment, each capacitor sample was tested one at a time after which the conventional capacitors were tested likewise. The ripple in the rectified signal was measured using a well-calibrated dual trace Cathode Ray Oscilloscope (CRO).

Figure 4. Experimental features for the performance evaluation

Each time a capacitor was to be replaced, the connecting leads from the power unit were disengaged from the circuit until the replacement was done as intended. In order to ensure that accurate readings were obtained, the capacitor under test was removed and the load resistor was also disengaged from the

circuit before the probes of the CRO were connected across the open end of the circuit to measure the ripple. After that, the resistor and capacitor were inserted in the circuit and the ripple in the rectified voltage reaching the resistor was measured. For the identical capacitor samples, the readings obtained were averaged and standard error was computed in each case. The ripple factor was also computed. In the case of each category of the fabricated capacitors, the mean capacitance value was used in the calculation of the ripple factor. The formula used in the computation was [39]

$$
V_r = V_{out}(1 - e^{-\frac{t}{RC}})
$$
 (2)

where V_r = peak ripple factor of the capacitor, V_{out} = peak value of the rectified signal less potential barrier of the diode used, $t =$ signal time (reciprocal of frequency, 50 Hz), $R =$ resistance of the load resistor (10 M Ω), and $C =$ capacitance of the capacitor. The silicon diode used in this study has potential barrier of 0.7 V.

The setup used for the determination of temperature coefficient of capacitance consisted of the electric hotplate (Model E4102 WH), cylindrical aluminium block, retort stand with clamp, and digital thermometer (Model 305, equipped with type –K probe). The capacitor sample to be tested was made to rest in the cavity of the aluminium block placed on the heating element of the hotplate. The tip of the thermometer probe was clamped, ensuring that it made a firm contact with one cross-section of the capacitor. The probes of the LCR meter were connected properly to the lead terminals of the capacitor for measurement of capacitance. These probes as well as the tip of the thermometer probe were properly and thickly lagged with cotton wool. This was necessary to ensure that heat radiation from the top surface of the aluminium block did not influence the measurements. Figure 5 shows the features of the measurement setup used. In this case, the hotplate served as the source of heat. By adjusting the control dial of the hotplate, heat supplied to the assembly was made to flow steadily through the block into the capacitor sample. As temperature of the sample increased by 5 $^{\circ}$ C from 20 $^{\circ}$ C, the capacitance value measured by the LCR meter was read until the thermometer registered 70° C. From the data obtained, the temperature coefficient of capacitance was calculated based on two standards, namely, Japanese Industrial Standards (JIS) and Electronic Industries Alliance (EIA). The JIS and EIA rely on 20 $^{\circ}$ C and 25 $^{\circ}$ C, respectively, as the reference temperature. The empirical relation applied for the calculation in this case was [40]

$$
\mathcal{C}_t = \left(\frac{c_m - c_r}{c_r \Delta T}\right) 100\,\%
$$
\n(3)

where C_t = temperature coefficient of capacitance, C_r = capacitance value at reference temperature, ΔT = difference between maximum operating temperature and reference temperature values, and C_m = capacitance value at operating temperature.

The relative permittivity was evaluated at various temperatures considered for the capacitor samples in this study. The relative permittivity value was obtained by applying the relation [41], [42], [43]

$$
C = \varepsilon_r \varepsilon_o \left(\frac{A}{d}\right) \tag{4}
$$

where $C = \text{mean}$ capacitance value per temperature, ε_r = relative permittivity of the capacitor samples, $A = \text{cross-sectional area of the sample } (1.767 \times 10^{-4} \text{ m}^2),$ $d =$ thickness of the sample, and $\varepsilon_o =$ permittivty of free space (8.85 x 10^{-12} Fm⁻¹).

Figure 5. Setup for Capacitance -Temperature Data Measurements

The relative permittivity values obtained for the capacitor samples were assessed statistically using Statistical Package for the Social Sciences (SPSS) Software. Specifically, oneway analysis of variance (ANOVA) was used to test the observed differences in the values at $p < 0.05$ (level of significance of 0.05) and Pearson's Product-Moment Correlation Coefficient (PPMCC) was determined for the obtained values.

4. Results and Discussion

The results of Carr's index measurements of the CEP and DCS are presented in Table 1. In any fabrication/manufacturing process, flowability is very important and it is inversely proportional to compressibility of powdery solids. A Carr's index that is less than 15 % signifies high flowability [44].

Now, since the CEP and DCS have Carr's indices that satisfy this condition, it can be adjudged that they are capable of ensuring very good content uniformity as the starting materials in the fabricated capacitor samples. By implication, the CEP and DCS are suitable for use as intended in this study. All the Carr's indices are approximately the same, meaning that the different ages of the eggshells used in preparing the CEP materials have no influence whatsoever on the flow behavior. This, probably, is due to the fact that the flow characteristic is dependent on the particle size of a powdery material.

Table 2 shows the capacitance of the capacitor samples at various temperatures. It is seen that the age of the eggshells has influence on the capacitance of the CEP-based capacitors. The CS05 has the highest capacitance, followed by the CS15 and then the CS30. Since chicken eggshell is $95 - 97$ % Calcium carbonate $(CaCO_3)$ crystals [45] and calcium is a metallic element, it could be that insulating ability of the shells decreases with storage time thus enabling the resulting samples to be more electrically-conductive. In this study, the CEP serves as dielectric. Das [46] posited that dielectrics are the materials which are poor in conducting electricity compared to metals which are very good conductors of electricity.

So, in the instant case, as the electrical conductivity tendency increases, the dielectric nature is reduced. It can be deciphered from the results that over the considered temperature range (0 \degree C to 70 \degree C), the capacitance decreases by about 48.26 %, 71.93 %, and 67.08 % for the CS05, CS15, and CS30 respectively. That is indicative of the effect temperature has on the capacitance of the CEP-based capacitors. The capacitance at 50 $^{\circ}$ C for the CS05 tends to compare well with the value obtained for the CS15 at 25 $^{\circ}$ C. Figure 6 reveals that, irrespective of the storage time (age) of the eggshells utilised, the capacitance of the produced capacitor declines progressively with temperature. Increased spacing between the trend representing the effect on CS05 and the case involving the CS15 signifies difference in stability. In other words, the CEP at age of 15 days contains more calcium than the one at age of 5 days as earlier averred. Since metals respond to temperature changes faster than nonmetals, there is a greater tendency for the CS15 to exhibit decrease in capacitance faster than the CS05 for the same interval of change in temperature.

Figure 6. Variation of capacitance with temperature of the capacitor samples

In Table 3, the temperature coefficients of capacitance of the capacitor samples are recorded based on the JIS and EIA standards. It is clear from the results that the fabricated capacitors have negative temperature coefficient of capacitance. This is possible because their capacitances decrease with increase in temperature. With respect to the reference temperature for each considered standard, it appears from the results that realization of very satisfactory performance would be possible within audio frequency range. The coefficients in this case compare favorably with the values ranging from -1.48 %/ $^{\circ}$ C to -0.97 %/ $^{\circ}$ C as reported by Umanah et al. [40] for disc-shaped compact fabricated from periwinkle shell powder.

Table 3. Temperature Coefficient of capacitance of the capacitor samples

Capacitor sample code	$C_{\rm t}$ (% / °C)		
	ЛS	EIA	
CS05	-0.97	-0.90	
CS15	-1.44	-1.39	
CS30	- 1.34	-1.34	

Like in the cases of capacitance, the relative permittivity values of the CEP-based capacitors depend on the storage time of the eggshells utilized. It can be seen from Table 4 that the CEP at 5 days gives rise to the highest value of the relative permittivity whereas the CEP at 30 days yields the lowest value. Sedha [47] reported that the relative permittivity of ceramic capacitors range from 6 to 7500. At all the temperatures considered in this work, the CS30 has relative permittivity that lies within this reported range. As

for the CS05 and CS15, values of relative permittivity lower than the maximum value as reported for ceramic capacitors are possible when the temperature is at least 35 $\mathrm{^{\circ}C}$ and 30 $\mathrm{^{\circ}C}$ respectively. Another factor that influences the relative permittivity of the CEP-based capacitors is temperature. For a change from 20 \degree C to 70 \degree C, the relative permittivity decreases by 48.27 %, 71.92 %, and 67.07 % in the cases of the CS05, CS15, and CS30 respectively.

Table 4. Relative Permittivity per temperature of the Capacitor samples

Temperature,		Relative permittivity, ε_r	
T ($^{\circ}$ C)	CS ₀₅	CS15	CS30
20.0	9137	7837	5719
25.0	7970	5863	4768
30.0	7633	5146	4267
35.0	7039	4553	3775
40.0	6824	4072	3315
45.0	6159	3530	2742
50.0	5924	3039	2486
55.0	5617	2824	2292
60.0	5136	2640	2067
65.0	4799	2415	1975
70.0	4727	2200	1883

Figure 7 depicts that the variation of the relative permittivity with temperature of the CEP-based capacitors is both inverse and non-linear. By comparing the values statistically using ANOVA and PPMCC, it is revealed (as presented in Table 5) that very high and strong positive correlations exist among the capacitor samples in terms of their relative permittivity values.

Figure 7. Relative permittivity versus temperature of the capacitor samples

Concerning the ANOVA employed to assess the differences in the obtained values of the relative permittivity, the critical *F*-value at $p < 0.05$ is 5.117 (From ANOVA table). Among the calculated *F-*values, 1.5311 only is less than this critical value. This observation implies that the relative permittivity of the CS15 and that of the CS30 do not differ significantly. This agrees with the highest correlation (99.24 %) obtained for the pair whereas pairing the CS05 with either CS15 or CS30 agrees by a lesser percentage (98.24 % or 98.93 % respectively).

Table 5. Results of statistical assessments of the relative permittivity values

Treatment S	Source	Sum of squares	Degree οf freedo	Mean sum of squares	Calculate d F-value	PPMC C
			m			
CS ₀₅	Betwee	3275944	1	3275944		
VS	$\mathbf n$	1		1	13.146	0.9824
CS15	Within	4983858	20	2491929		
		8				
	Total	8259802	21			
		9				
CS ₀₅	Betwee	5785349	1	5785349		
VS	$\mathbf n$	8		8	31.927	0.9893
CS30	Within	3624143	20	1812071		
		7				
	Total	9409493	21			
		6				
CS15	Betwee	3544040	1	3544040		
VS	$\mathbf n$				1.5311	0.9924
CS30	Within	4629401	20	2314700		
		\mathfrak{D}				
	Total	4983805	21			
		3				

From Table 6, it is seen that the corresponding conventional capacitors for comparison of performances have approximately the same capacitance value. Also, the differences in the ripple factors when comparing the CS05, CS15, and CS30 with their conventional ceramic counterparts are 4.89 %, 2.04 %, and 1.64 % respectively. These values are very insignificant and thus negligible. The computed values of the ripple factor are obviously negligible as well especially when comparing the pairs of the data obtained between the use of capacitance of the conventional ceramic capacitors and that of the CEP-based capacitor samples. The closeness in values revealed in these cases indicate that the capacitor samples developed from the eggshells have comparative advantage and could serve as suitable alternatives to the conventional ceramic capacitors.

Values in parenthesis are based on capacitances of the conventional ceramic capacitors

5. Conclusion and Future Scope

Findings from the experimental investigations conducted in this study have shown that chicken eggshell powder has Carr's index of about 9.0 %, indicating an acceptable flow characteristic for manufacturing purpose. Capacitance (1.84 $nF - 8.93$ nF), relative permittivity (1883 – 9137), and temperature coefficient of capacitance $(-1.44 \text{ %} / {}^{o}C - 0.90)$ $%^{\circ}C$) of the capacitor samples varied with the age of the

eggshells used. The performances of the capacitor samples (reduction in ripples from 3.22 % - 8.00 %) compared favorably with those of conventional ceramic capacitors with similar capacitance values. Recycling chicken eggshells, as described in this study, could help to ensure availability of cost-effective capacitors and valorization of the eggshells serves as a scientifically-safe way of solving the associated disposal problems. For further research in this direction, eggshell treatment methods and their effects on the capacitor characteristics should be investigated for possible optimization of capacitor manufacturing processes.

Data Availability

All data are with the corresponding author.

Conflict of Interest

The authors declare that they have no conflict of interests regarding this paper's publication.

Funding source

No explicit support was received for this study from the public, corporate or non-profit organizations.

Authors' Contributions

Levi Oye: Created the methodology for the research, analyzed the data, and wrote the first draft of the manuscript.

Nsikak Edet Ekpenyong: Contributed to the final version's permission for submission, gave critical feedback on the manuscript's structure and contents, and served as the lead supervisor.

Aniefiok Otu Akpan: Reviewed and edited the document to make sure it was coherent and very clear, serves as the cosupervisor, helped to proofread and approve the submission of the finished version.

Acknowledgements

None

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