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Towards fractional-order capacitors with broad tunable constant phase angles: multi-walled carbon nanotube-polymer composite as a case study

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ABSTRACT

In this study, multi-walled carbon nanotube (MWCNT) filled Polyvinylidene fluoride-trifluoroethylene-chlorofluoroethylene (PVDF-TrFE-CFE) composites are used to realize fractional-order capacitors (FOCs). A solution-mixing and drop-casting approach is used to fabricate the composite. Due to the high aspect ratio of MWCNTs, percolation regime starts at a small weight percentage (wt%), 1.00 %. The distributed MWCNTs inside the polymer act as an electrical network of micro-capacitors and micro-resistors, which, in effect, behaves like a FOC. The resulting FOCs' constant phase angle (CPA) can be tuned from -65° to -7° by changing the wt% of the MWCNTs. This is the largest dynamic range reported so far at the frequency range from 150 kHz to 2 MHz for an FOC. Furthermore, the CPA and pseudo-capacitance are shown to be practically stable (with less than 1% variation) when the applied voltage is, changed between 500 μ V and 5V. For a fixed value of CPA, the pseudo-capacitance can be tuned by changing the thickness of the composite, which can be done in a straightforward manner via the solution-mixing and drop-casting fabrication approach. Finally, it is shown that the frequency of a Hartley oscillator built using an FOC is almost 15 times higher than that of a Hartley oscillator built using a conventional capacitor.

1. Introduction

The capacitor is a fundamental electrical device consisting of an insulator sandwiched between two metal electrodes and is generally used to store and control the charge and energy in circuit [1, 2]. An ideal insulator does not support any conduction current, and the (conventional) capacitor introduces a -90° phase difference between the voltage and current measured at its electrode [2, 3]. Controlling this phase angle as an additional degree of freedom permits design of electrical circuits that would not be possible using conventional capacitor [4-7]. One of way of achieving this is through the use of fractional order capacitors (FOCs) with controllable phase angles [8-11]. Indeed, it has been shown that FOCs with constant phase angles (CPAs) that can be tuned in a large dynamic range have become indispensable in design and realization of more robust electrical system [12-14]. For example, replacing the conventional capacitor with an FOC reduces the power consumption and increases the reliability of analog proportional–integral–derivative (PID) controllers [15]. Another example is the modeling of spiking properties of neurons, where FOCs offer better accuracy than conventional passive circuit elements by introducing the capability of accounting for capacitive memory effects[16]. Last example is Hartley oscillators, where the generated oscillation frequencies are significantly increased by replacing conventional capacitors with FOC [17].

Although various realizations of FOCs have been developed so far, most of them suffer from one or more of the following shortcomings: bulkiness, microelectronic incompatibility, lack of tunability of CPA, low dynamic range of CPA, significant variation of CPA, narrow constant phase zone (CPZ—frequency range where CPA is obtained), high power consumption, and high fabrication cost [18-25]. Recently, a single-component FOC, where the insulator is made of Polyvinylidene fluoride–trifluoroethylene–chlorofluoroethylene (PVDF-TrFE-CFE) terpolymer containing reduced graphene oxide (rGO) nano-sheets, has been reported [9]. By changing the weight percentage (wt%) of rGO nanosheets from 2 to 12 in the composite, this FOC's CPA can be tuned from -65° to -31° in the CPZ ranging from 50 kHz to 2 MHz. However, the dual-step synthesizing process of rGO and the large amount of rGO increase the cost of FOC fabrication. In [10] an FOC, where the insulator is made of epoxy resin containing multi-walled carbon nanotubes (MWCNTs), has been proposed. It has been shown in the same work that this FOC's CPA can be tuned from -85° to -45° by increasing the wt% of MWCNTs from 0.75 to 3. This small amount of MWCNTs reduces the fabrication cost but the minimum CPA is limited to -45° . In addition, when the amount of MWCNTs is increased, FOC's CPZ shifts in frequency. It is important to note here that none of the FOC implementations described or cited above offer low fabrication cost or a tunable CPA with high dynamic range in fixed CPZ.

In this work, a new class of FOCs, where the insulator is ferroelectric PVDF-TrFE-CFE terpolymer containing MWCNTs, is developed. Fabrication cost is reduced by directly mixing MWCNTs and PVDF-TrFE-CFE and using a smaller percentage of MWCNTs. Changing the wt% of MWCNTs from 1 to 4.5 tunes the CPA in the large dynamic range from -65° to -7° in the fixed CPZ between 150 kHz and 2 MHz. The high aspect ratio of the MWCNTs helps to obtain this large dynamic range that can be controlled using a smaller amount of MWCNTs. The concept of implementing an FOC using MWCNT/PVDF-TrFE-CFE composite is illustrated in figure 1. The composite [shown in blue in figure 1(a)] is sandwiched between two gold electrodes [shown in yellow in figure 1(a)]. A magnified cross-section of the composite is shown in figure 1(b), where the black and blue rods represent the MWCNTs and PVDF-TrFE-CFE, respectively. “Electrical interactions” between MWCNTs observed in the percolation regime, which include electron transport (via hopping) and ohmic contact, and capacitive effect are schematized in figure 1(c). These interactions can be modeled in the form of an electrical network of micro-capacitors and micro-resistors as shown in figure 1(d) [26]. Since the dimension of each MWCNT is different and they are not distributed uniformly, the structure of the resulting network is rather complex with different values of capacitance and resistance. The response of this network has a fractional order behavior with a CPA in a wide CPZ.

2. Methods

To prepare the composite, 200 mg PVDF-TrFE-CFE is dissolved in a 2ml solvent, N, N-Dimethylformamide (DMF), under constant stirring at room temperature for two days. 2, 5, 7, 8, 8.5, and 9 mg of MWCNTs (corresponding to wt% of 1.00, 2.50, 3.50, 4.00, 4.25, and 4.50) are suspended in 1ml DMF, and dispersed via ultrasonication for 1 hour. The dispersed MWCNT solutions are poured onto the dissolved PVDF-TrFE-CFE polymer solution and mixed under continuous stirring for another 24 hours. It should be noted here that the minimum wt % is selected to be 1.00 since it has been reported previously that the percolation starts for values of wt% higher than 0.75 [10]. The average diameter and average lateral length of the MWCNTs are 200 nm and 10 μ m, respectively, corresponding to an average aspect ratio of 50). Au-covered, 2 cm $\times$ 2 cm Si/SiO₂ wafers are used to fabricate the FOC by drop-casting the composite solution. 10 nm Ti and 190 nm Au are deposited on Si/SiO₂ wafers via DC sputtering to construct the bottom electrode. The composite solutions are drop-casted and dried for 12 hours at 85 $^\circ$ C under a vacuum. The top electrode is a circular Au film with 3mm diameter and 200nm thickness deposited in a similar way using a shadow mask. This approach gives us the freedom to fabricate nine individual FOCs on a 2 cm $\times$ 2 cm sample area. The sample is flip-bonded on a PCB board, so that each capacitor provides a separate connection for the electrical measurements.

A 2 cm × 2 cm chip with nine individual capacitors, which is fabricated as described above, is shown in figure 2(a). The MWCNTs are clearly distinguished from the polymer in the scanning electron microscopy (SEM) cross-section of the composite provided in figure 2(b). Furthermore, X-ray diffraction (XRD) patterns of the composite with 4.5 wt% MWCNT and only the PVDF-TrFE-CFE film are compared in figure 2(c). Note that the XRD spectra are normalized with respect to the gold peak at 38°. XRD spectrum of the film shows an intense peak at 18.2° (corresponding to a (111) plane). As expected, the composite also shows a peak at 18.2° but its amplitude is smaller. An additional peak is observed at 26.1° in the spectrum of the composite due to interlayered spacing between adjacent graphite layers of MWCNTs. The fact that composite's XRD spectrum does not show additional peaks confirms that no additional complex molecular structures formed in the composite.

3. Results and Discussion

Agilent 4980A LCR meter is used to measure the magnitude of the impedance $|Z|$ and phase angle ϕ of the individual FOCs in the frequency range between 50 kHz to 2 MHz. The amplitude of the applied voltage is 500 mV and thickness of composites are 30 μm unless stated otherwise.

Figure 3(a) plots ϕ versus frequency for different values of the MWCNT wt %. Figure clearly shows that ϕ stays nearly constant in the frequency range between 150 kHz and the 2 MHz; this range is identified as the CPZ for all FOCs with different wt%. It should be noted here that CPZ overlaps with the frequency range where dipolar relaxation occurs in the ferroelectric polymer [27, 28]. Additionally, the CPAs of the FOCs with wt% of 1.00, 2.50, 3.50, 4.00, 4.25, and 4.50 are found to be -65° , -53° , -47° , -30° and -7° respectively, by averaging the values of ϕ samples in the CPZ. Figure 3(b) plots the normalized histogram of the difference between phase angle of the samples (for all six FOCs) and the CPAs and shows that 88% of the samples are within $\pm 3^\circ$ of the CPAs. Normalization is done with respect to total number of measured points. Figure 3(a) and the CPA values clearly show that the CPA increases with the wt% of MWCNTs and approaches zero. Larger amount of MWCNTs inside the polymer means more ohmic contact generating higher number of paths to support conduction current.

For an FOC, the magnitude of the impedance can expressed as $|Z| = 1/(C_a \omega^\alpha)$, where C_a is pseudo-capacitance (sometimes also called fractance) and α ($0 < \alpha < 1$) is the fractional order [17]. Fractional order α of each capacitor is calculated by applying (linear) least-squares fit to the samples of $\log|Z|$ in the CPZ. Note that

1 $\log|Z| = -\alpha \log \omega - \log C_\alpha$. The values of α are found to be 0.73, 0.59, 0.53, 0.39, 0.30 and 0.09 for FOCs with wt%
 2 1.00, 2.50, 3.50, 4.00, 4.25, and 4.50, respectively [figure 3(c)]. The maximum relative error (with respect to least-
 3 squares fit value) in the measured samples are 0.02, 0.01, 0.02, 0.035, 0.03 and 0.02 respectively. Note that the above
 4 α values correspond to the CPA values of -65.7° , -53.1° , -47.7° , -35.1° , -27.1° , and -8.1° and confirm the CPA
 5 values obtained by averaging.

6 Figure 4(a) plots $C_\alpha = 1/(|Z|\omega^\alpha)$ versus frequency, where values of $|Z|$ are the measured samples and values
 7 of α are those computed using least-squares fit as described above. The figure clearly shows that C_α is nearly
 8 constant in the CPZ. The average value of C_α increases from $2 \text{ nF sec}^{-0.27}$ to $50 \text{ mF sec}^{-0.91}$ as the wt% of the MWCNTs
 9 is increased from 1.00 to 4.50. This is expected since the composite with a larger amount of MWCNT has a higher
 10 dielectric constant. It should be noted here that (average) C_α depends also on the surface area and thickness of
 11 composite film. Figure 4(b) plots ϕ and C_α versus frequency for the FOCs with 2.5 wt% of the MWCNTs and three
 12 different values of film thickness and demonstrates that response of the FOC can be tuned by changing the geometrical
 13 dimensions. It is important to mention that we observed a resistive behavior in the composite films with thickness 10
 14 μm and $15 \mu\text{m}$. This can be explained as the distributed MWCNTs make a conductive path between electrodes since
 15 the average length of MWCNT is $10 \mu\text{m}$.

16 Next, the effect of applied voltage on the CPA and (average) C_α is characterized. Figure 5(a) plots ϕ versus
 17 frequency for the FOC with 2.5 wt% of the MWCNTs under different values of the applied voltage changing between
 18 $500 \mu\text{V}$ and 5 V . The figure shows that ϕ is nearly constant (around the CPA of -55°) in the CPZ for all values of the
 19 applied voltage. The inset of figure 5(a) plots the CPA values (estimated by averaging the values of ϕ samples as
 20 described above) versus the applied potential. The maximum change in the CPA is found to be only 0.2° . Similarly,
 21 figure 5(b) plots C_α versus frequency for the same FOC under the same values of the applied voltage and shows that
 22 it stays nearly constant in the CPZ for all values of the applied voltage. The inset of figure 5(b) plots the average C_α
 23 versus the applied potential. The maximum change is found to be only $5 \times 10^{-9} \text{ F s}^{-0.4}$ (2.5%). These results confirm
 24 that the response of the fabricated FOCs is stable for values of the applied voltage changing between $500 \mu\text{V}$ and 5
 25 V.

To demonstrate the advantage of an FOC over a conventional capacitor, oscillation frequencies of two Hartley oscillators [figure 6(a)], one using an FOC and another one using a conventional capacitor are compared. The oscillation angular frequency ω_{osc} satisfies the following equation:

$$\omega_{osc}^{(1+\alpha)} - \frac{2r+R}{L} \omega_{osc}^{\alpha} - \frac{2}{LC_{\alpha}} \sin\left(\frac{\alpha\pi}{2}\right) = 0 \quad (1)$$

where R is the resistance of the resistor and L and r are the inductance and internal resistance of the inductors, respectively. Note that setting $\alpha = 1$ in (1) gives the oscillation frequency ($f_{osc} = \frac{\omega_{osc}}{2\pi}$) of the Hartley oscillator with the conventional capacitor. First, the dependence of f_{osc} on α is characterized assuming $C_{\alpha} = 50 \text{ nFs}^{(\alpha-1)}$. Equation (1) is solved for f_{osc} for values of α changing between 0.1 and 1. Note that this corresponds to CPA changing from -90° to -10° . Figure 6(b) plots f_{osc} versus α , which show that the frequency of oscillation could be increased significantly with slightly decreasing the α . Next, Hartley oscillator using the FOC fabricated with 2.5 wt% MWCNT (corresponding to $\alpha = 0.6$ and $C_{\alpha} = 50 \text{ nFs}^{-0.4}$) is built. Figure 6(c) plots the response of the oscillator, demonstrating that the oscillation frequency is 178 kHz. Figure 6(d) plots the response of the Hartley oscillator built using the conventional capacitor with $\alpha = 1.0$ and $C_{\alpha} = 50 \text{ nF}$ demonstrating that the oscillation frequency is 11.75 kHz. These results show that the oscillation frequency of the Harvey oscillator with the fabricated FOC is almost 15 times of that of the Harvey oscillator with the conventional capacitor. This demonstrates the suitability of FOCs for high frequency applications. It should be noted here that solving equation (1) for $\alpha = 0.6$ and $\alpha = 1$ yields oscillation frequencies of 186.12 kHz and 11.98 kHz, respectively. The discrepancy between the measured and calculated values can be explained as deviation in the α , where the oscillation frequency is sensitive to α as given in figure 6(b).

4. Conclusions

A new FOC, where the insulator is made of an MWCNT/PVDF-TrFE-CFE composite, is developed. The fabrication cost of this new FOC is expected to be lower than that of the previously developed FOC, which uses a rGO/PVDF-TrFE-CFE composite [9]. This is simply because the fabrication process in this work employs a simple solution-mixing and drop-casting approach and the wt% of the MWCNTs is smaller than the wt% of the rGO. The new FOCs' CPA has a larger dynamic range (obtained by changing the wt% of the MWCNTs) than the CPA of the

previously developed FOC, which uses an MWCNT/Epoxy composite [10]. Additionally, the new FOC's CPZ does not shift in frequency as the wt% of the MWCNT is changed to tune the CPA. Indeed, results presented in this work demonstrate that the CPA of the new FOC can be tuned in the broad range between -65° and -7° in the fixed CPZ between 150 kHz and 2 MHz. To the best of author's knowledge, this is the largest dynamic range that has been reported for the CPA. Additionally, the results demonstrate that the change in the applied voltage (between 500 μ V to 5 V) has almost no effect on the characteristics of the FOC. Finally, it is shown that the frequency of a Hartley oscillator built using this new FOC is almost 15 times higher than that of a Hartley oscillator built using a conventional capacitor. All in all, this work demonstrates that FOCs fabricated using CNT-ferroelectric polymers composites have the potential to become essential components for reliable/robust electrical and electronic systems.

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Figures

Figure 1. Concept. (a) 3D schematics of the FOC. Top and bottom electrodes of the FOC are shown in yellow. (b) The MWCNT/PVDF-TrFE-CFE composite. Black and blue rods represent the MWCNTs and the PVDF-TrFE-CFE terpolymer, respectively. (c) Illustration of the network of micro-capacitors (C) and micro-resistors (R), ohmic contact, and electron transport and the chemical formula of PVDF-TrFE-CFE. (d) Approximate network electrically equivalent to the composite. The network impedance behaves as an FOC.

Figure 2. (a) 2 cm $\times$ 2 cm fabricated chip area with nine FOCs. (b) Cross-sectional SEM image of MWCNT/PVDF-TRFE-CFE composite. The MWCNT with bright contrasts forms a microcircuit as depicted in figure 1. (c) XRD spectra for PVDF-TrFE-CFE and MWCNT/PVDF-TrFE-CFE. XRD spectra. Note that the spectra are normalized with respect to the main Au (111) peak at 38.3°. The first inset zooms to 2θ range between 15° and 22° while the second one zooms to 25°-30° range. The peaks in the first and second insets belong to PVDF-TrFE-CFE and MWCNTs, respectively.

Figure 3. (a) Phase angle versus frequency between 150 kHz and 2 MHz for the FOCs with different MWCNT wt %. Dotted lines represent the CPA obtained by averaging the phase angle samples. (b) Normalized histogram of the phase angle difference between measured and fitted values for all six FOCs (c) Impedance magnitude versus frequency in logarithmic scale. The slope in the equation represent the α , and the results confirm the CPAs shown in figure 3(a).

Figure 4. (a) Pseudo-capacitance versus frequency between 150 kHz and 2 MHz for the FOCs with different MWCNT wt %. (b) Phase angle and pseudo-capacitance versus frequency for the FOC with 2.5 wt% MWCNT and various thickness.

Figure 5. (a) Phase angle versus frequency between 150 kHz and 2 MHz for the FOC with 2.5 wt% MWCNT under different values of applied voltage. The inset shows the CPA versus applied voltage between $500 \mu\text{V}$ and 5 V . (b) Pseudo-capacitance versus frequency. The inset shows the mean pseudo-capacitance versus applied voltage.

Figure 6. Fractional order Hartley sinusoidal oscillator. (a) Schematic and realization. (b) Oscillation frequency versus α . Output voltage (c) for $\alpha = 0.6$ and (d) for $\alpha = 1$.

References

- [1] B. Chu, X. Zhou, K. Ren, B. Neese, M. Lin, Q. Wang, F. Bauer, Q. M. Zhang, *A dielectric polymer with high electric energy density and fast discharge speed*, Science 313(5785) (2006) 334-336.
- [2] J. Ho, T.R. Jow, S. Boggs, *Historical introduction to capacitor technology*, IEEE Electrical Insulation Magazine 26(1) (2010) 20-25.
- [3] A.K. Jonscher, *Dielectric relaxation in solids*, Journal of Physics D: Applied Physics 32(14) (1999) R57.
- [4] M.D. Ortigueira, *An introduction to the fractional continuous-time linear systems*, IEEE Circuits and Systems Magazine 8(3) (2008) 19-26.
- [5] A.S. Elwakil, *Fractional-order circuits and systems: an emerging interdisciplinary research area*, IEEE Circuits and Systems Magazine 10(4) (2010) 40-50.
- [6] C.A. Monje, Y.Q. Chen, B.M. Vinagre, D.Xue, V. Feliu-Battle, *Fractional-order Systems and Controls: Fundamentals and Applications*, Springer, 2010.
- [7] K. Biswas, G. Bohannan, R. Caponetto, A.M. Lopes, J.A.T. Machado, *Fractional-Order Devices*, Springer (2017).
- [8] A. Agambayev, S. Patole, M. Farhat, A. Elwakil, H. Bagci, K.N. Salama, *Ferroelectric Fractional-Order Capacitors*, ChemElectroChem, 4(11), (2017), 2807-2813.
- [9] A.M. Elshurafa, M.N. Almadhoun, K.N. Salama, H.N. Alshareef, *Microscale electrostatic fractional capacitors using reduced graphene oxide percolated polymer composites*, Applied Physics Letters 102(23) (2013) 232901.
- [10] D. John, S. Banerjee, G.W. Bohannan, K. Biswas, *Solid-state fractional capacitor using MWCNT-epoxy nanocomposite*, Applied Physics Letters 110(16) (2017).
- [11] A.K. Gil'mutdinov, P.A. Ushakov, R. Khazali, *Fractal Elements and their Applications*, Springer, (2017).
- [12] A.G. Radwan, K.N. Salama, *Fractional-order RC and RL circuits*, Circuits, Systems, and Signal Processing 31(6) (2012) 1901-1915.
- [13] A. Radwan, A. Shamim, K.N. Salama, *Theory of fractional order elements based impedance matching networks*, IEEE Microwave and Wireless Component Letters 21(3) (2011) 120-122.
- [14] K. Moaddy, A.G. Radwan, K.N. Salama, S. Momani, I. Hashim, *The fractional-order modeling and synchronization of electrically coupled neuron systems*, Computers & Mathematics with Applications 64(10) (2012) 3329-3339.
- [15] G.W. Bohannan, *Analog fractional order controller in temperature and motor control applications*, Journal of Vibration and Control 14(9-10) (2008) 1487-1498.
- [16] S.H. Weinberg, *Membrane capacitive memory alters spiking in neurons described by the fractional-order Hodgkin-Huxley model.*, PloS one 10(5) (2015) e0126629.
- [17] A. Elwakil, A. Agambayev, A. Allagui, K.N. Salama, *Experimental demonstration of fractional-order oscillators of orders 2.6 and 2.7*, Chaos, Solitons & Fractals 96 (2017) 160-164.
- [18] S. Roy, *On the realization of a constant-argument immittance or fractional operator*, IEEE Transactions on Circuit Theory 14(3) (1967) 264-274.
- [19] K. Biswas, S. Sen, P.K. Dutta, *Realization of a constant phase element and its performance study in a differentiator circuit*, IEEE Transactions on Circuits and Systems II: Express Briefs 53(9) (2006) 802-806.

[20] T.C. Haba, G. Ablart, T. Camps, F. Olivie, *Influence of the electrical parameters on the input impedance of a fractal structure realised on silicon*, Chaos, Solitons & Fractals 24(2) (2005) 479-490.

[21] J. Valsa, J. Vlach, *RC models of a constant phase element*, International Journal of Circuit Theory and Applications 41(1) (2013) 59-67.

[22] A. Agambayev, S. Patole, H. Bagci, K.N. Salama, *Tunable fractional-order capacitor using layered ferroelectric polymers*, AIP Advances 7(9) (2017) 095202.

[23] G. Tsirimokou, C. Psychalinos, A. Elwakil, K. Salama, *Experimental verification of on-chip CMOS fractional-order capacitor emulators*, Electronics Letters, 52(15) (2016) 1298-1300.

[24] G. Tsirimokou, C. Psychalinos, A.S. Elwakil, K.N. Salama, *Electronically Tunable Fully Integrated Fractional-Order Resonator*, IEEE Transactions on Circuits and Systems II: Express Briefs (2017).

[25] M.S. Sarafraz, M.S. Tavazoei, *Passive Realization of Fractional-Order Impedances by a Fractional Element and RLC Components: Conditions and Procedure*, IEEE Transactions on Circuits and Systems I: Regular Papers 64(3) (2017) 585-595.

[26] C.W. Nan, Y. Shen, J. Ma, *Physical properties of composites near percolation*, Annual Review of Materials Research 40 (2010) 131-151.

[27] Y. Wang, S.G. Lu, M. Lanagan, Q. Zhang, *Dielectric relaxation of relaxor ferroelectric P(VDF-TrFE-CFE) terpolymer over broad frequency range*, IEEE transactions on ultrasonics, ferroelectrics, and frequency control 56(3) (2009).

[28] L. Zhu, *Exploring strategies for high dielectric constant and low loss polymer dielectrics*, The journal of physical chemistry letters 5(21) (2014) 3677-3687.