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Multi-material Fused Deposition Modelling for integration of interdigital dielectric sensors into carbon fibre composite tooling for in process cure monitoring

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Abstract

This study introduces the concept of printing two thermoplastic materials in a single 3D Printing process – one conductive the other non-conductive – to form interdigital sensor structures for Dielectric Analysis in mould tools for cure monitoring of the epoxy matrix during the Carbon Fibre Reinforced Polymer manufacturing process. This low-cost method would allow for reduced assembly and ease of sensor placement directly from Computed Aided Design, whilst using an accessible technology.

The results obtained showed a cure point can be identified in accordance with the epoxy specifications, via integration of interdigital sensors manufactured using conductive composite PolyLactic Acid with multi-material Fused Deposition Modelling.

Keywords: fused deposition modelling; multi-material; smart tooling; cure monitoring; composites; resin infusion

Funding: This work was supported by the Aerospace Technology Institute (ATI) Project 113036, Horizon AM.

1. Introduction

Carbon Fibre Reinforced Polymer (CFRP) is a composite material comprising of woven carbon fibres within an epoxy polymer matrix, for applications which require a material with a high specific strength [1,2]. Production methods include open and closed mould methods such as Liquid Resin Infusion (LRI) and Resin Transfer Moulding (RTM), respectively. The use of these materials in high quality assurance industries such as Aerospace [3] present the demand for a high level of in-process monitoring and control. The ability to track the cure progress of the polymer matrix avoids the de-moulding of under-cured CFRP components, exhibiting poor physical and chemical properties [4].

Non-destructive monitoring methods enable the assessment of the CFRP cure progress without impacting the functionality of the finished part. Many have been explored to date, including the use of optical fibres [5], ultrasonic [6], inductive coupling [7] and dielectric [4,8-13] methods. The limiting factor for these technologies is the capital costs of sensors, monitoring equipment and occasionally the need to place sensors in invasive positions (i.e. within the final component [10] or on the mould tool to dispose of afterwards).

Curing of an epoxy results in an increase in the ion viscosity of the material [6, 12]. Dielectric Analysis (DEA) monitors the changing dielectric properties of the Material Under Test (MUT) separating two electrodes; one with an alternating current (AC) applied to it, with the other acting as a sensing electrode [12]. The complex expression of dielectric properties, ϵ^* is shown in Equation 1.

$$\epsilon^* = \epsilon' - j\epsilon'' \tag{1}$$

where ϵ' is the relative permittivity and ϵ'' is the dielectric loss factor.

The dielectric constant is the ability of the MUT to store energy and diminishes as the ion viscosity increases. When the AC signal is applied, an alternating electric field is generated between the two electrodes; orientating the ions within the epoxy MUT to the changing field direction (Figure 1).

As the cure progresses, the mobility of ions is restricted, reducing the dielectric constant of the MUT, a part of parallel plate capacitance, C (F) in Equation 2 - therefore capacitance decreases as the epoxy cure progresses until a plateau is observed when polymerisation is complete [12].

$$C = \epsilon' A / d \tag{2}$$

where A (m^2) is the surface area of the parallel plates and d (m) is the distance between them.

A parallel plate setup presents an option for DEA for closed mould production of CFRP parts with simple geometry [13]. However, ensuring consistent spacing and alignment between electrodes is challenging, particularly when using open mould methods such as LRI; which do not require a top plate to seal the mould as with RTM. Interdigital Sensors (IDS) present an advantage in this case, as the co-planar electrodes maintain relatively constant positions to each other [4, 12], and can be placed directly onto the mould tool surface regardless of geometry complexity.

Emergence of commercial multi-material Fused Deposition Modelling (FDM) machines – allowing for two separate materials to be extruded in a single print [14-15] – alongside the development and application of multifunctional composite conductive thermoplastics [16-19], make this technology an option for printing IDS dielectric sensors directly into the mould tool, presenting several advantages. The IDS are located at accurately predefined positions using Computer Aided Design (CAD), allowing for sensor placement at areas of concern or interest. There are no additional assembly steps for sensor placement at the layup stage. Sensor placement on potentially difficult features such as a compound curved edge is simple.

It is hypothesised that IDS geometry can be directly manufactured into mould tools for LRI by using conductive composite thermoplastic in a multi-material FDM process. The printed sensors could prove an effective method for performing DEA to asses epoxy curing progress.

Using this technology, IDS sensors were directly manufactured into test coupons using multi-material FDM to extrude a composite conductive PLA alongside a standard insulating PLA. These test coupons were used to monitor the curing process of an infusion epoxy resin and identify a complete cure point. A representative mould tool was manufactured for testing using LRI.

2. Manufacturing Process

The test samples were manufactured on a multi-material FDM system. Two different filaments were extruded through separate nozzles. Nozzle 1 extruded a standard silver coloured PLA and Nozzle 2 extruded a conductive composite PLA (ProtoPasta) to achieve the final embedded sensor designs shown in Figure 2. The final sensor geometry in Figure 2 was driven from experiments to avoid short-circuits via nozzle ooze during travel moments. This would be detrimental to the capacitive effect used in DEA. The A to D naming convention for each IDS as in Figure 2 will remain consistent throughout all figures.

3. Experimentation

The experimental setup consisted of a computer connected to an Inductance-Capacitance-Resistance (LCR) monitor via a General Purpose Interface Bus (GPIB) controller. The DUT was connected to the LCR, with an AC signal applied to obtain capacitance readings without polarising the epoxy dielectric. A frequency sweep list (1 kHz, 10 kHz and 100 kHz) was setup on the LCR monitor with a manual trigger to obtain readings at each frequency. The trigger was controlled by a MATLAB script to capture a reading every 10 seconds.

A two-part commercially available infusion epoxy resin was used for the cure tests, with the manufacturer's specification quoting gelation at 2 to 4 hours, a de-mould time within 6 to 8 hours and cure time of 24 hours at 25°C with final properties achieved in 7 days. Table 1 shows the mass of epoxy used for each sensor design (note that the arc electrodes in Figure 2d required a greater mass in order to cover the entire electrode surface) and the base capacitance of each sensor (reading with no epoxy present over the sensor). It is important to consider the mass of epoxy in each coupon, to account for thermal runaway effects resulting in faster cure times.

Each sensor model was produced in batches of 3 with all undergoing experimentation with the results showing minimal intra and inter-batch variation.

4. Results

4.1. Epoxy cure monitoring through capacitance

Figure 3 shows the effect of the progression of epoxy cure on the capacitance of each of the printed dielectric sensors. The magnitude of capacitance increases inversely to frequency due to polarisation of the electrodes at lower frequencies [12].

A reading of the ambient air was taken prior to pouring in the epoxy sample, which caused a significant increase in the observed capacitance. At this stage, the ions and dipoles in the epoxy are free to orientate to the alternating field direction generated by the LCR monitor – hence they contribute to the energy storage capacity of the sensor. From this point the curing process begins, increasing the mechanical and ion viscosity – causing a rapid decrease in the capacitance of the sensor within the first 10 hours. It is noted that there is a consistent change in the rate at which capacitance falls at 1 kHz in Figure3a-d at approximately 2 hours; potentially the onset of gelation in accordance with the epoxy specification. The capacitance then proceeds to a plateau, where the ions and dipoles in the epoxy are restricted and therefore cannot store energy via orientation to the field direction. Figure 3a-c shows that the addition of insulating layers over the sensor results in a reduction in sensitivity, with the overall percentage change from peak to plateau being 37.9%, 28.07% and 12.27% respectively. The curved sensor in Figure 3d follows the same trend, but achieves a saturation point earlier than the planar examples caused by the larger mass of epoxy having a thermal runaway effect.

4.2. Definition of epoxy cure point

The cure point of the epoxy is when cross-linking has completed and its properties are constant, which can be ascertained by analysing the percentage change hour-over-hour of the sensor capacitance. Figure 4 shows the reduction in the rate of change of capacitance until the noise level of the LCR monitor is reached when there is a change of sign from negative to positive.

Figures 4a-c show a change of sign close to 24, 25 and 24 hours respectively after the onset of the curing process; a useful indicator of the conclusion of polymerisation, it is not a reversible process – the noise level of the LCR monitor has been reached. This holds true for Figure 4(d) after 21 hours – a faster cure time likely due to thermal runaway caused by a larger mass of epoxy as discussed.

4.3. Example mould tool with embedded IDS

To demonstrate the application of this study, a mould tool with an IDS printed into the surface to monitor the cure progress of an LRI process (Figure 5). The mould tool was prepared with carbon fibre fabric, peal ply, inlet/outlet connectors and enclosed in a vacuum bag. The outlet connector was connected to a vacuum pump, with the inlet connector in a cup with epoxy – drawing it through the carbon fibre. The bag was then sealed, the vacuum pump switched off and the capacitance of the integrated IDS was monitored over a period of 30 hours.

Figure 6a shows the capacitance change over the observed period. From observation, there is a large spike in capacitance during the infusion process, separated out into Figure 6b for clarity, with Figure 6d showing the same time period from the surface electrode test coupon shown in Figure 2a for comparison. This anomaly is caused by the vacuum pump drawing the epoxy over the sensor – resulting in a mobile dielectric medium – and subsides when the pump was switched off at 6 minutes (0.1 hours).

Figure 6c enables the identification of a cure point using the method discussed in 4.2. The first change of sign at 27 hours puts the cure point beyond those of the test coupons. A longer cure is expected due to a smaller volume of epoxy being present over the IDS when compared to previous examples, as the exothermic reaction dissipates heat faster than a larger mass in an epoxy pot – reducing the speed of the cure.

In order to supplement the identified cure point in Figure 6c, a separate LRI process was carried out. The CFRP coupon was de-moulded at 26.5 hours – just prior to the point identified in Figure 6c – and a digital durometer was used to obtain the Shore D hardness readings shown in Table 2. The manufacturers specification quotes a Shore D range of 86 to 90 at 25 °C. The measured values in Table 2 show a Shore D range of 87.5 to 91.5 – achieving the epoxy cure specification.

Conclusions

IDS geometry can be printed directly into mould tools fabricated using a multi-material FDM process with conductive composite PLA. The manufactured sensors can be used for epoxy cure monitoring via DEA for applications in production of CFRP. The obtained results show that a cure point can be identified by observing the percentage change of capacitance hour-over-hour; consistent with manufacturer times and Shore D hardness specifications. Adding insulating layers to conceal IDS electrodes results in reduced sensitivity, but still has functionality for DEA.

Direct FDM printing of these sensor structures in a multi-material process allows for ease of use in open mould processes such as LRI, and potentially has further applications for optimization of cure cycles [20] and flow analysis on-the-fly [13]. It also presents several advantages including: definition of locations in CAD, no assembly required, ease of placement onto complex geometry and low cost of manufacture whilst using a widely accessible technology.

Acknowledgements

The authors would like to acknowledge funding from the Aerospace Technology Institute (ATI) Project 113036, Horizon AM. The authors also acknowledge Dr R Watson for assistance in setting up laboratory equipment, and the University of Warwick's Engineering Build Space (School of Engineering) for the use of their equipment during this study.

References

1. Hughes R R, Drinkwater B W, Smith R A. Characterisation of carbon fibre-reinforced polymer composites through radio-transform analysis of complex eddy-current data. Composites Part B: Engineering 2018;148:252-259.
2. Sharma M, Gao S, Mäder E, Sharma H, Wei L W, Bijwe J. Carbon fiber surfaces and composite interphases. Composites Science and Technology 2014;102(1):35-50.
3. Fawaz, Z. Quality control and testing methods for advanced composite materials in aerospace engineering. Advanced Composite Materials for Aerospace Engineering 2016;429-451.
4. Yang, Y et al. Development of a dielectric sensor system for the on-line cure monitoring of composites. Procedia Technology 2014; 15:631-637.
5. Wang P, Li D, Shang Q, Chen Z, Wang H. Cure monitoring of epoxy resin via use of FBG. 7th International Forum on Strategic Technology 2012;1-4.
6. Pommer C, Sinapius M. A novel approach to monitoring the curing of epoxy in closed tools by use of ultrasonic spectroscopy. Sensors (Basel) 2018; 18(1):96.
7. Chilles J S, Koutsomitopoulou A F, Croxford A J, Bond I P. Monitoring cure and detecting damage in composites with inductively coupled embedded sensors. Composites Science and Technology 2016; 134:81-88.
8. Skordos A A, Karkanas P I, Partridge I K. A dielectric sensor for measuring flow in resin transfer moulding. Measurement Science and Technology 1999;11:25-31.

9. Raponi O A, Raponi R A, Barban G B, Benedetto R M D, Ancelotti A C. Development of a simple dielectric analysis module for online cure monitoring of a commercial epoxy resin formulation. Materials Research 2017;20(2):291-297.
10. Boll D, Schubert K, Brauner C, Lang W. Miniaturized flexible interdigital sensor for in situ dielectric cure monitoring of composite materials. IEEE Sensors Journal 2014;14(7):2193-2197.
11. Mcllhagger A, Brown D, Hill B. The development of a dielectric system for the on-line cure monitoring of the resin transfer moulding process. Composites Part A 2000; 31(12):1373-1381.
12. Yang Y, Chiesura G, Vervust T, Degrieck J, Vanfleteren J. Design and fabrication of a shielded interdigital sensor for noninvasive in situ real-time production monitoring of polymers. Journal of Polymer Science Part B Polymer Physics 2016; 54(20).
13. Yenilmez B, Murat Sozer E. A grid of dielectric sensors to monitor mold filling and resin cure in resin transfer molding. Composites Part A: Applied Science and Manufacturing 2009;40(4):476-489.
14. Yin J, Lu C, Fu J, Huang Y, Zheng Y. Interfacial bonding during multi-material fused deposition modelling (FDM) process due to inter-molecular diffusions. Materials and Design 2018;150:104-112.
15. Kim H, Park E, Kim S, Park B, Kim N, Lee S. Experimental study of mechanical properties of single- and dual-material 3D printed products. Procedia Manufacturing 2017;10:887-897.
16. Leigh S J, Bradley R J, Purssell C P, Billson D R, Hutchins D A. A simple, low-cost conductive composite material for 3D printing of electronic sensors. PLoS One 2012;7(11):e49365.
17. Flowers P F, Reyes C, Ye S, Kim M J, Wiley B J. 3D printing electronic components and circuits with conductive thermoplastic filament. Additive Manufacturing 2017; 18: 156-163.
18. Katseli V, Economou A, Kokkinos C. Single-step fabrication of an integrated 3D-printed device for electrochemical sensing applications. Electrochemistry Communications 2019; 103:100-103.
19. S W Kwok et al. Electrically conductive filament for 3D-printed circuits and sensors. Applied Materials Today 2017; 9:167-175.
20. de Andrade Raponi O, Righetti de Souza B, Miranda Barbosa L C, Ancelotti Junior, A C. Thermal, rheological, and dielectric analyses of the polymerization reaction of a liquid thermoplastic resin for infusion manufacturing of composite materials. Polymer Testing 2018; 71:32-37.

Vitae

Elliott Griffiths is a Mechanical Engineer currently working toward his PhD in the Digital and Material Technologies Laboratory (DMTL) at the University of Warwick. His research area involves hybridisation of additive manufacturing processes for incorporation of embedded electronics – materials, methods and applications.

Dr Simon Leigh is an Associate Professor of Engineering at the University of Warwick, where he leads the Digital and Material Technologies Laboratory. His research is undertaken in the field of Additive Manufacturing (AM, also known as 3D printing) and focusses on the development and application of novel materials and processes for high-resolution functional AM and multi-material AM.

Figure Captions

Figure 1. *Parallel plate setup with separating dielectric epoxy material (a) ions free to move with no applied electric field (b) ions polarised with applied AC to generate electric field at time instance.*

Figure 2. *IDS dielectric sensor coupons manufactured using multi-material FDM (a) Sample A IDS with surface electrodes, (b) Sample B IDS with electrodes covered by 0.2 mm layer of PLA, (c) Sample C IDS with electrodes covered by two 0.2 mm layers (0.4 mm total) of PLA and (d) Sample D IDS with surface electrodes on an example radius.*

Figure 3. *Graphs showing the change of capacitance over time and frequency across (a) Sample A IDS with surface electrodes, (b) Sample B IDS with electrodes covered by a 0.2 mm layer of PLA, (c) Sample C IDS with electrodes covered by two 0.2 mm layers (0.4 mm total) of PLA and (d) Sample D IDS with surface electrodes on an example radius.*

Figure 4. *Graphs showing the percentage change of capacitance hour-over-hour with frequency across (a) Sample A IDS with surface electrodes, (b) Sample B IDS with electrodes covered by a 0.2 mm layer of PLA, (c) Sample C IDS with electrodes covered by two 0.2 mm layers (0.4 mm total) of PLA and (d) Sample D IDS with surface electrodes on an example radius. Red lines show point of changing sign, where a cure point has been identified.*

Figure 5. *FDM mould tool with IDS printed into the surface for cure monitoring in the LRI process.*

Figure 6. *Graphs showing (a) the change of capacitance over time and frequency across IDS with on surface of FDM mould tool over 30 hours (b) percentage change of capacitance hour-over-hour with frequency across (c) change of capacitance over first 30 minutes and (d) the first 30 minutes on the surface electrodes from Sample A for comparison.*

Tables

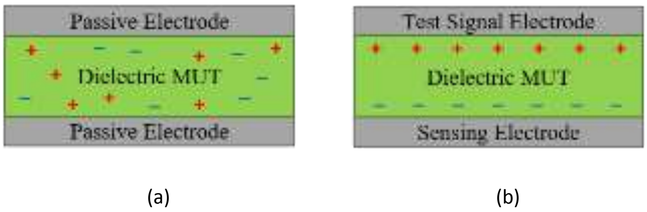
	Sample A	Sample B	Sample C	Sample D
Epoxy mass (g)	10.5	9.1	11.0	19.3
Capacitance in air (pF) @ 10 kHz	15.2	15.2	20.6	17.7

Table 1 *Table of epoxy mass and initial capacitance readings.*

	Sample Position A	Sample Position B	Sample Position C	Sample Position D	Sample Position E	Sample Position F	Average
Shore D Hardness	91	90	87.5	88	91.5	91	89.8

Table 2 *Table of measured Shore D Hardness readings taken over the cured CFRP component de-moulded after 26.5 hours cure time.*

Figures



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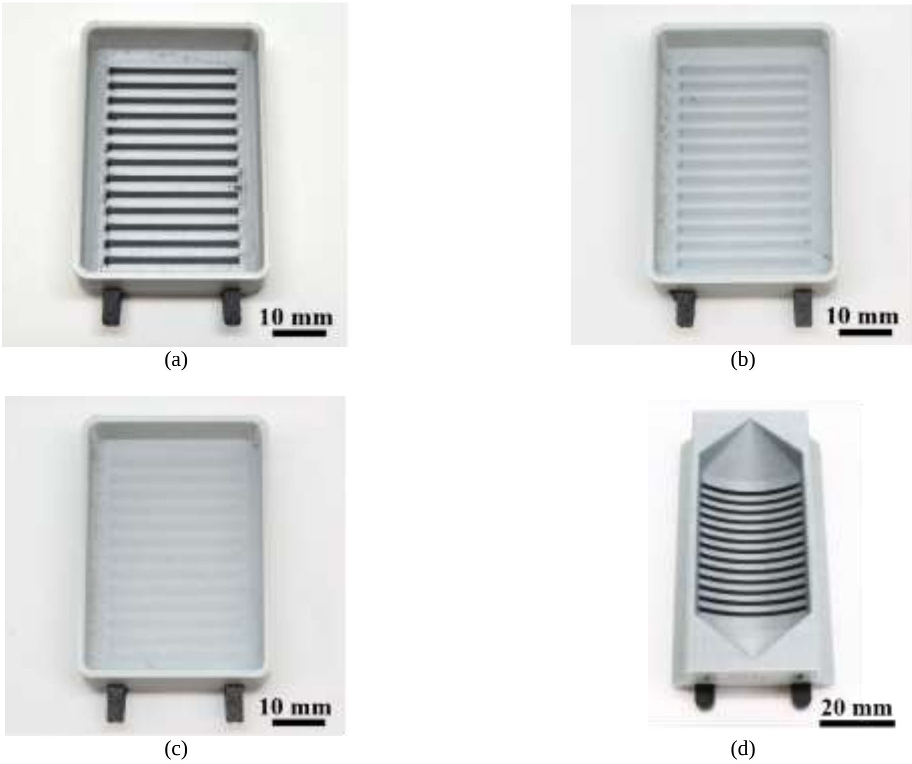

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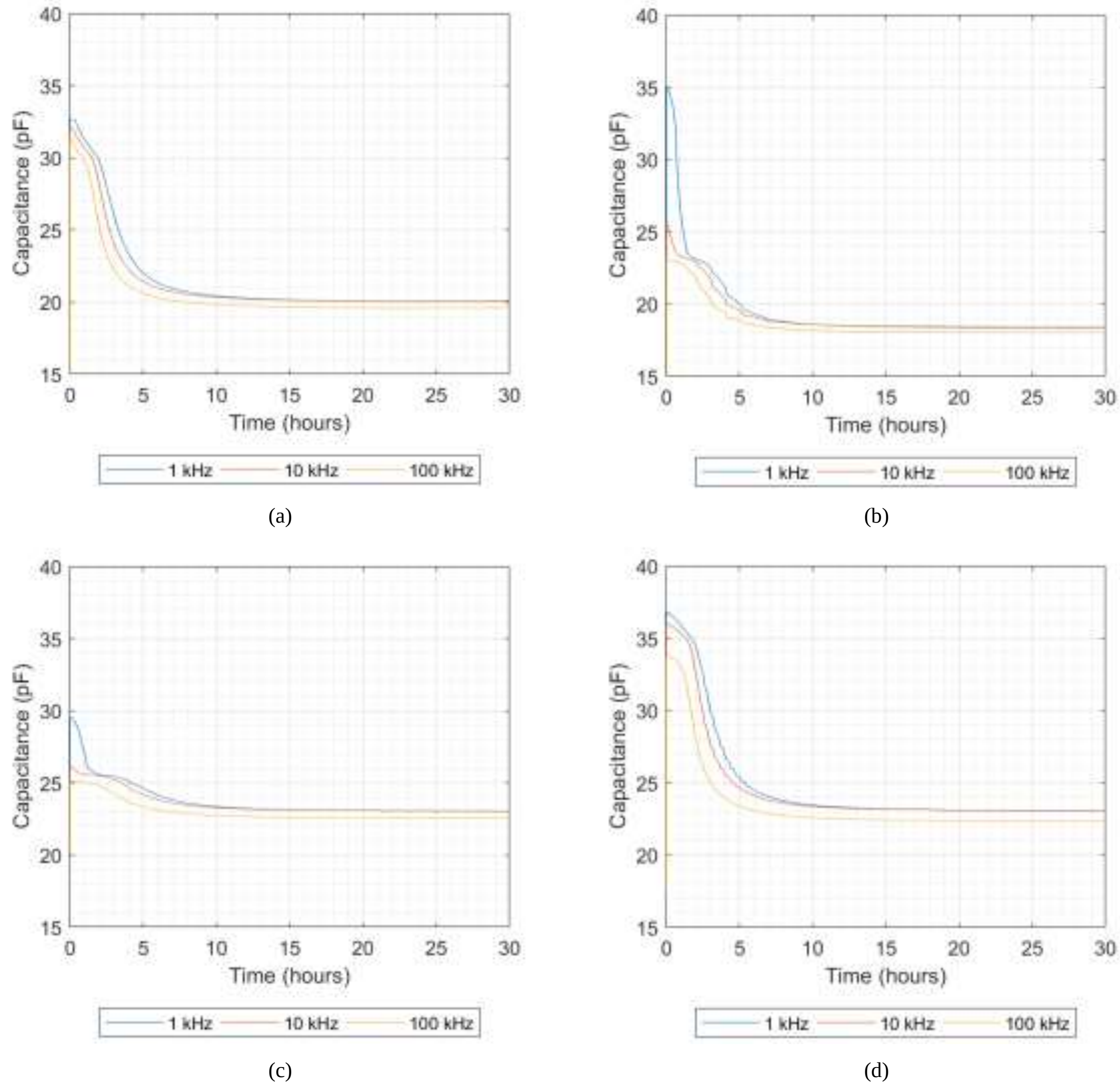

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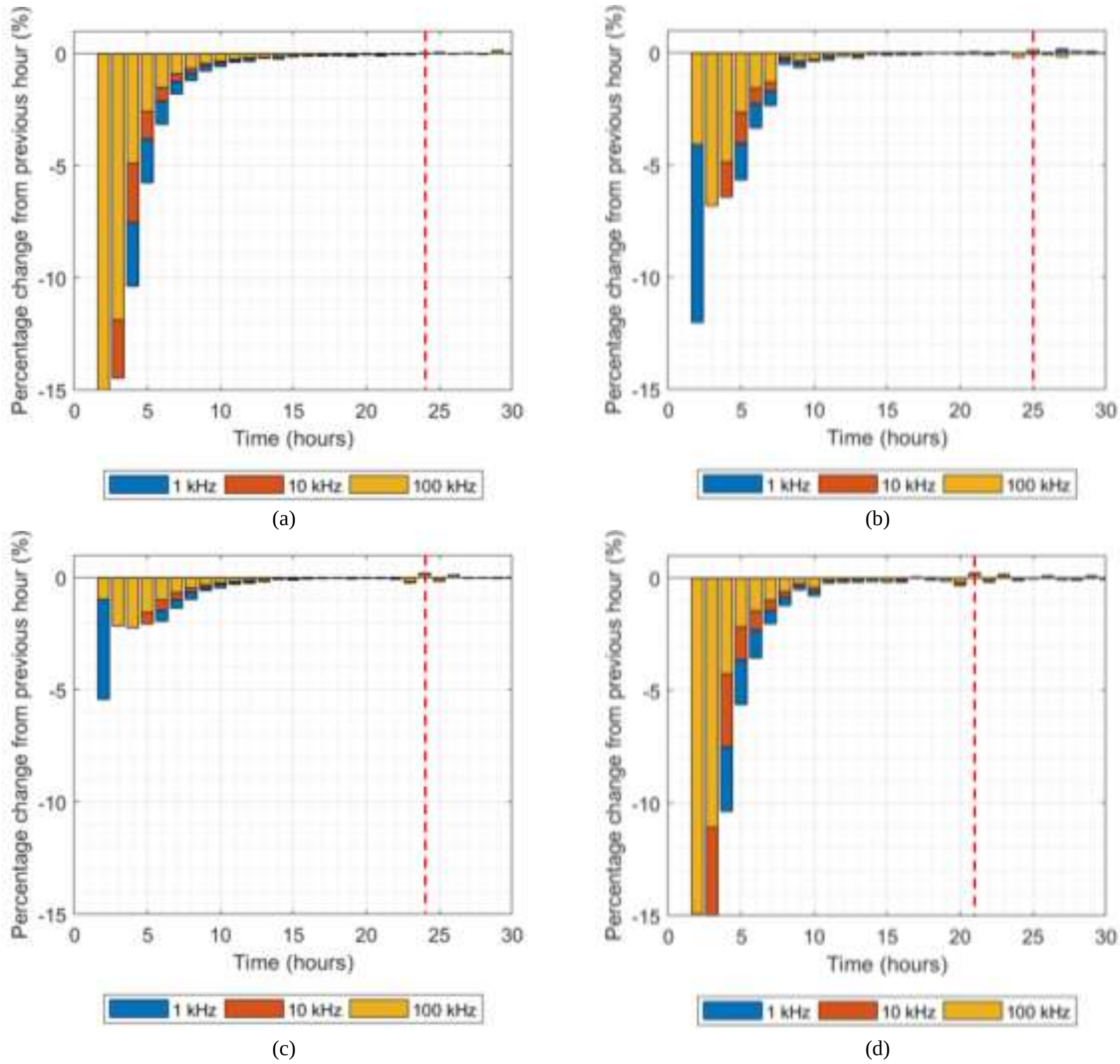

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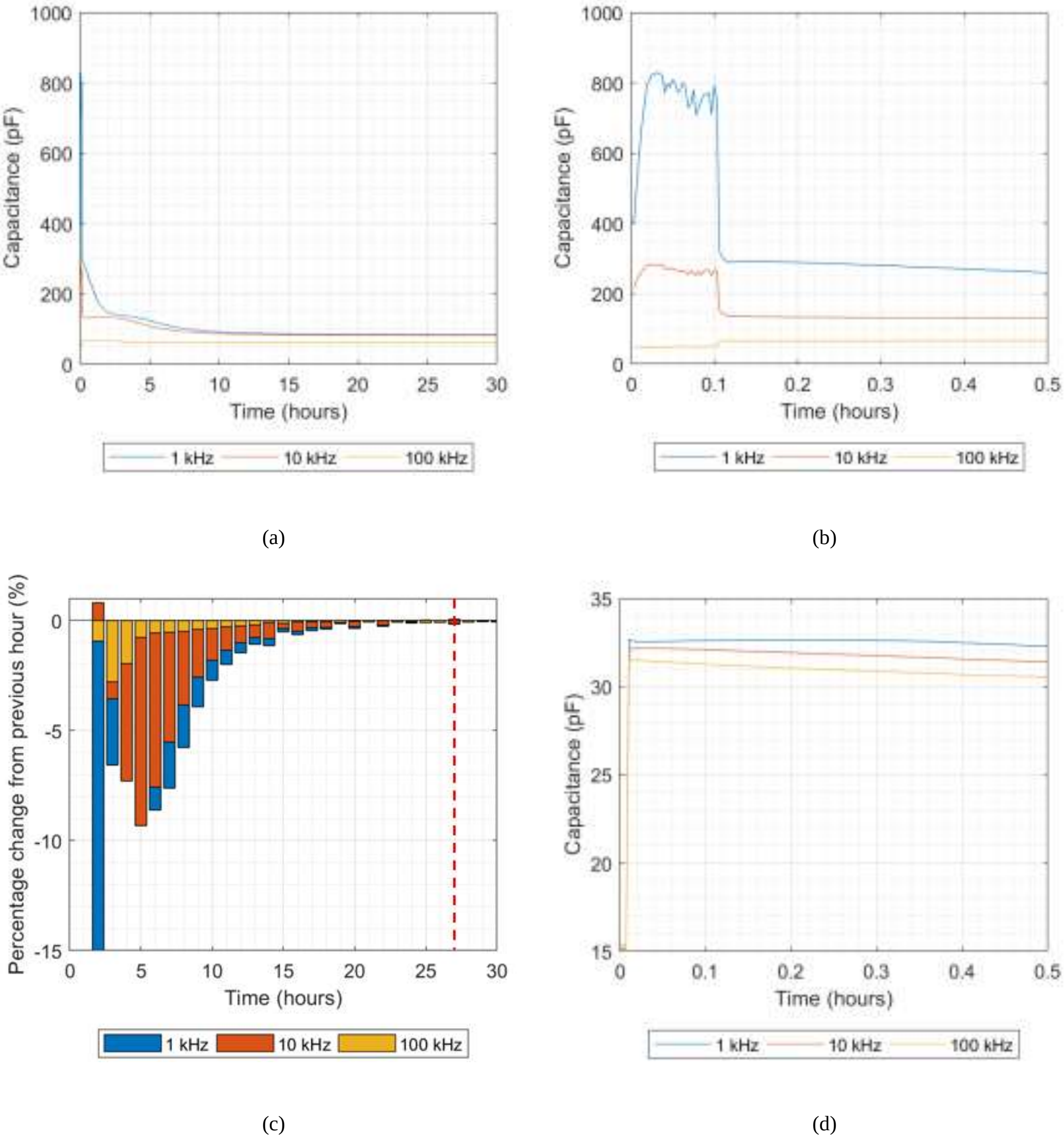

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