

Layer-by-layer printing: how we fabricate the next generation of nanocomposite capacitors for more efficient power electronics

William Greenbank¹*, Shova Neupane¹, Bartosz Gackowski¹, Luciana Tavares¹, Thomas Ebel¹

1. Centre for Industrial Electronics, University of Southern Denmark, Alsion 2, DK-6400, Denmark

* presenting/corresponding author: greenbank@sdu.dk

Further reading

For details on the layer-by-layer nanocomposite printing technique outlined in this summary, the reader is referred to:

Greenbank, W., & Ebel, T. (2023). Layer-by-layer printable nano-scale polypropylene for precise control of nanocomposite capacitor dielectric morphologies in metallised film capacitors. *Power Electronic Devices and Components*, 4, 100025.

Context

Electricity generation accounts for 47% of all new carbon emissions because electricity production is expected to increase by 80% by 2040 – a significant portion from fossil fuel sources.[1], [2] It is therefore necessary to both curb rising demand for energy and increase renewable energy's share of electricity generation to have any realistic hope of reducing emissions long-term. More efficient power electronics can have an enormous impact on energy wastage. Capacitors are critical to the operation of power electronics, but often the weak link when it comes to efficiency improvements.[3], [4] This is particularly true for electric motors, which account for 40% of all global electricity consumption and this will only increase as electric vehicles become more prevalent.[5] Reducing energy waste in motors requires that their drives are smaller and can tolerate higher temperatures while remaining highly reliable and stable at high voltages.[6] However, existing dielectric materials cannot deliver a capacitor that meets all of these requirements.

Overview of nanocomposite capacitors

In the context of power capacitors, nanocomposite dielectrics are typically envisaged as a polymer matrix/continuous phase containing one or more ceramic nanomaterials (materials with at least 1 dimension in the 1 – 100 nm length scale).[7] A representation of this and some of the different material possibilities are shown in Figure 1.

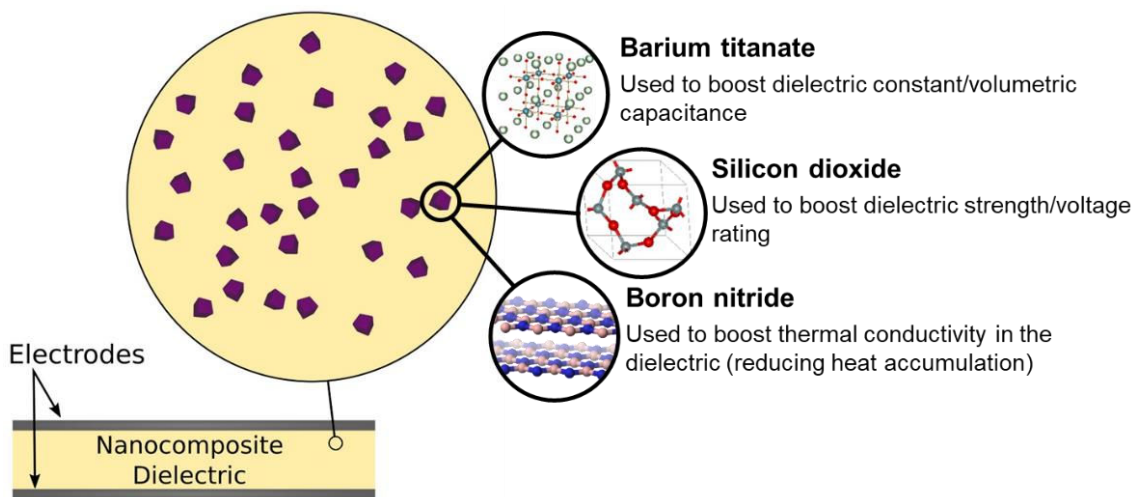


Figure 1: an illustration of a nanocomposite dielectric with some examples of nano-dopant materials and their uses.[8]

Different nanomaterials may be used to enhance different dielectric properties, such as high- ϵ_r materials to boost the volumetric/specific capacitance of the device,[8] high dielectric strength materials to boost breakdown voltage,[9] thermally conductive materials to remove heat from the device,[10] and conductive materials to reduce dissipation factor.[11] While individually effective, these nano-dopants are most effective when used together in multi-material systems, such as sandwich dielectrics[12]–[14] which include alternating layers doped with either high- ϵ_r nanoparticles or high-dielectric strength nanoparticles, in order that the best combination of dielectric properties can be achieved.

Challenges in nanocomposite capacitor fabrication

Such a seemingly simple solution to dielectric material engineering should already be well established in the capacitor market, however commercial nanocomposite capacitors are practically non-existent. This is because numerous challenges exist in their fabrication. When placed in a polymer matrix, ceramic nanoparticles have a tendency to cluster, driven by a high interfacial energy between the particle and the polymer and therefore a thermodynamic driving force towards reducing the area of the interface by clustering.[15]

The formation of nanoparticle clusters in a nanocomposite dielectric has several important consequences. First is that the dielectric properties of the particles do not transfer well to the nanocomposite,[8] but perhaps more important is that such clusters can act as defect sites for dielectric breakdown.[16], [17] The maximum energy density of a parallel-plate capacitor is given in equation (1):

$$U = \frac{1}{2} \epsilon_0 \epsilon_r E^2 \quad (1)$$

In which U is the maximum energy density (per unit volume) and E is the breakdown field strength (voltage/unit thickness). Therefore, any potential gain in energy density is completely undone if clustering causes even a slight decline in breakdown voltage.

Furthermore, nanomaterials pose significant challenges for device fabrication. The vast majority of metallised film capacitors are fabricated by extruding and stretching the dielectric polymer from a melt (e.g., biaxially oriented polypropylene - BOPP).[18], [19] Nanoparticles added to a hot polymer melt have decreased kinetic stabilisation owing to the heat of the melt, increasing the likelihood of clustering, which can decrease the quality of the extruded film.[20] Such clusters also act as fracture initiation sites during stretching, limiting the extent to which stretching can be used to decrease film thickness and orient the polymer.[21]

Layer-by-layer nanocomposite dielectric fabrication

At the University of Southern Denmark Centre for Industrial Electronics we have developed a layer-by-layer printing method for fabricating nanocomposite dielectrics for capacitors (Figure 2(1-4)).[22] This technique will utilise standard industrial-scale printing methods, such as roll-to-roll slot die coating, to deposit layers of polymer or polymer/nanocomposite dielectrics utilising novel polymer gel inks based on industry-friendly solvents.[22] In addition to offering unparalleled thickness and morphology control, this technique permits the commercial scale production of numerous different device architectures which would be impractical or impossible using extrusion-based fabrication (Figure 2(5-7)).[22] This includes the “sandwich dielectric” which is favoured by researchers for the implementation of nanocomposite dielectrics.[12]–[14]

A series of prototype devices were printed with this technique based on polypropylene. The devices printed without the inclusion of nanoparticles exhibited predictable and reproducible capacitances and thicknesses, with dielectric properties comparable with commercial polypropylene.[22] Sandwich-like devices were also fabricated with alternating layers of pure polypropylene and polypropylene doped with barium titanate nanoparticles. These devices exhibited a 20% improvement in dielectric constant with no significant decline in breakdown voltage with the addition of only 0.77% (v/v) BaTiO₃ nanoparticles.[22] This is in good agreement with recent theoretical findings, which indicate that even greater improvements in dielectric constant are attainable without sacrificing breakdown performance.[23]–[25]

Future perspectives

Layer-by-layer dielectric printing is an enabling technique that opens the door to a range of new research pathways and commercial capacitor design possibilities. With this technology, manufacturers will have the ability to design nanocomposite dielectric morphologies for specific applications. Moreover, the controlled fabrication of reliable nanocomposite capacitors will allow the promises of more compact, high temperature stable capacitors to be realised, decreasing the size of motor drives and facilitating the transition to high-efficiency wide band-gap semiconductor-based power electronics in e-mobility applications.

One thing that has been emphasised in this work and others is the importance of nanoparticle surface engineering, both from the perspective of preventing nanoparticle clustering and in promoting good transfer of dielectric properties from the dopant ceramic to the nanocomposite.[8], [26] This is an area of ongoing research that is expected to bear fruit in the near future. In addition to nanoparticles, the possibility of using other additives, such as cross-linking agents to improve the thermal resiliency of the layer is made possible with this technique, as well as the systematic study of the effects of different nano-dopants in a controlled/systematic fashion in an industry-relevant setting. Finally, the scalability of this technique will be explored, with the first trials of commercial-scale printing anticipated in 2024.

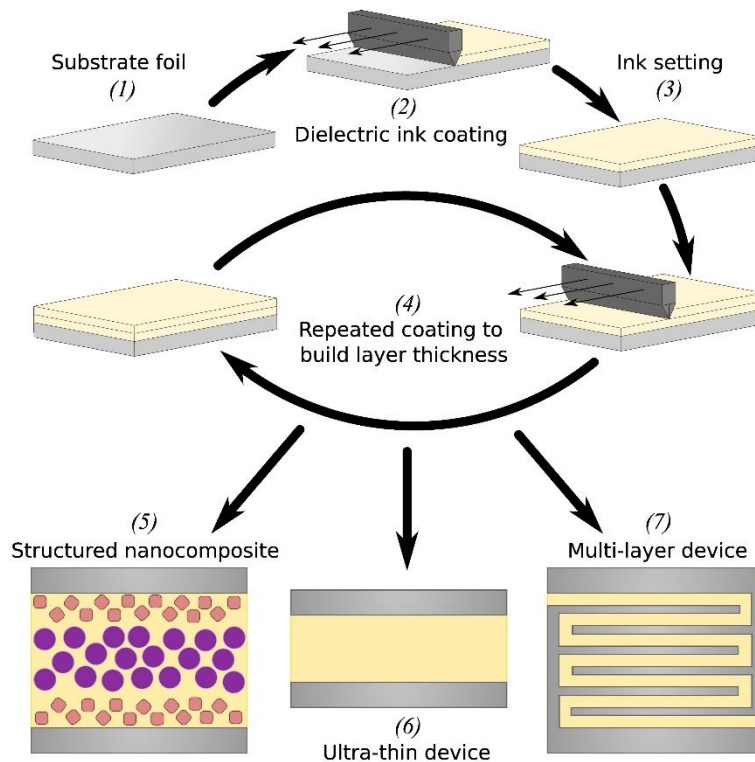


Figure 2: (1-4) the layer-by-layer nanocomposite dielectric printing process developed at the University of Southern Denmark and (5-7) some of the device architectures made possible with this technique.

References

- [1] R. K. Pachauri and L. A. Meyer, "Climate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change," Geneva, 2014.
- [2] "International Energy Agency World Energy Outlook 2014," Paris, 2014. [Online]. Available: <https://www.iea.org/reports/world-energy-outlook-2014>
- [3] J. Watson and G. Castro, "A review of high-temperature electronics technology and applications," *Journal of Materials Science: Materials in Electronics*, vol. 26, no. 12, pp. 9226–9235, Dec. 2015, doi: 10.1007/s10854-015-3459-4.
- [4] H. Jedtberg, M. Langwasser, R. Zhu, G. Buticchi, T. Ebel, and M. Liserre, "Impacts of unbalanced grid voltages on lifetime of DC-link capacitors of back-to-back converters in wind turbines with doubly-fed induction generators," in *2017 IEEE Applied Power Electronics Conference and Exposition (APEC)*, Tampa: IEEE, Mar. 2017, pp. 816–823. doi: 10.1109/APEC.2017.7930790.
- [5] R. Guerrero-Lemus and J. M. Martínez-Duart, *Energy Harvesting and Energy Efficiency*, vol. 37. in Lecture Notes in Energy, vol. 37. Cham: Springer International Publishing, 2017. doi: 10.1007/978-3-319-49875-1.
- [6] T. M. Jahns, "The Past, Present, and Future of Power Electronics Integration Technology in Motor Drives," *CPSS Transactions on Power Electronics and Applications*, vol. 2, no. 3, pp. 197–216, Sep. 2017, doi: 10.24295/CPSSSTPEA.2017.00019.
- [7] J. K. Nelson, Ed., *Dielectric Polymer Nanocomposites*. Boston, MA: Springer US, 2010. doi: 10.1007/978-1-4419-1591-7.
- [8] M. Streibl, R. Karmazin, and R. Moos, "Materials and applications of polymer films for power capacitors with special respect to nanocomposites," *IEEE Transactions on Dielectrics and Electrical Insulation*, vol. 25, no. 6, pp. 2429–2442, Dec. 2018, doi: 10.1109/TDEI.2018.007392.
- [9] I. Rytöluoto, K. Lahti, M. Ritamäki, and M. Karttunen, "The Role of Film Processing in the Large-Area Dielectric Breakdown Performance of Nano-Silica-BOPP Films," *Proceedings of the Nordic Insulation Symposium*, no. 24, Sep. 2017, doi: 10.5324/nordis.v0i24.2286.
- [10] Y. Abdullahi Hassan and H. Hu, "Current status of polymer nanocomposite dielectrics for high-temperature applications,"

Compos Part A Appl Sci Manuf, vol. 138, no. July, p. 106064, Nov. 2020, doi: 10.1016/j.compositesa.2020.106064.

- [11] J. Lu, K. S. Moon, and C. P. Wong, "Silver/polymer nanocomposite as a high-k polymer matrix for dielectric composites with improved dielectric performance," *J Mater Chem*, vol. 18, no. 40, pp. 4821–4826, 2008, doi: 10.1039/b807566b.
- [12] Y. Wang *et al.*, "Ultrahigh energy density and greatly enhanced discharged efficiency of sandwich-structured polymer nanocomposites with optimized spatial organization," *Nano Energy*, vol. 44, pp. 364–370, Feb. 2018, doi: 10.1016/j.nanoen.2017.12.018.
- [13] Q. Li *et al.*, "Sandwich-structured polymer nanocomposites with high energy density and great charge–discharge efficiency at elevated temperatures," *Proceedings of the National Academy of Sciences*, vol. 113, no. 36, pp. 9995–10000, Sep. 2016, doi: 10.1073/pnas.1603792113.
- [14] Z. Shi, J. Wang, F. Mao, C. Yang, C. Zhang, and R. Fan, "Significantly improved dielectric performances of sandwich-structured polymer composites induced by alternating positive-k and negative-k layers," *J Mater Chem A Mater*, vol. 5, no. 28, pp. 14575–14582, 2017, doi: 10.1039/C7TA03403B.
- [15] L. Song *et al.*, "Nanoparticle Clustering and Viscoelastic Properties of Polymer Nanocomposites with Non-Attractive Polymer-Nanoparticle Interactions," *Macromolecules*, vol. 55, no. 17, pp. 7626–7636, Sep. 2022, doi: 10.1021/acs.macromol.2c00689.
- [16] M. Saleem *et al.*, "Percolation phenomena of dielectric permittivity of a microwave-sintered BaTiO₃–Ag nanocomposite for high energy capacitor," *J Alloys Compd*, vol. 822, 2020, doi: 10.1016/j.jallcom.2019.153525.
- [17] J. Y. Li, L. Zhang, and S. Ducharme, "Electric energy density of dielectric nanocomposites," *Appl Phys Lett*, vol. 90, no. 13, p. 132901, Mar. 2007, doi: 10.1063/1.2716847.
- [18] J. L. Nash, "Biaxially oriented polypropylene film in power capacitors," *Polym Eng Sci*, vol. 28, no. 13, pp. 862–870, Jul. 1988, doi: 10.1002/pen.760281307.
- [19] D. M. Zogbi, "Global Power Capacitor Markets in 2020: Expect Slow and Steady Market Growth | TTI, Inc.," 2020. <https://www.tti.com/content/ttiinc/en/resources/marketeye/categories/passives/me-zogbi-20200309.html> (accessed May 12, 2021).
- [20] G. Filippone and D. Acierno, "Nanoparticle Dynamics in Polymer Melts," in *Smart Nanoparticles Technology*, InTech, 2012. doi: 10.5772/33647.
- [21] T. L. Anderson, *Fracture mechanics: fundamentals and applications*, 3rd ed. CRC press, 2005.
- [22] W. Greenbank and T. Ebel, "Layer-by-layer printable nano-scale polypropylene for precise control of nanocomposite capacitor dielectric morphologies in metallised film capacitors," *Power Electronic Devices and Components*, vol. 4, p. 100025, Mar. 2023, doi: 10.1016/j.pedc.2022.100025.
- [23] V. Bordo and T. Ebel, "Theory of Electrical Breakdown in a Nanocomposite Capacitor," *Applied Sciences*, vol. 12, no. 11, p. 5669, Jun. 2022, doi: 10.3390/app12115669.
- [24] V. Bordo and T. Ebel, "How to determine the capacitance of a nanocomposite capacitor," *AIP Adv*, vol. 12, no. 4, p. 045107, Apr. 2022, doi: 10.1063/5.0085619.
- [25] V. G. Bordo and T. Ebel, "The role of electron extinction in the breakdown strength of nanocomposite capacitors," *AIP Adv*, vol. 13, no. 6, Jun. 2023, doi: 10.1063/5.0150213.
- [26] J. Shao, M. Ji, and X. Liu, "A Developed Rahaman Model for Estimating the Dielectric Permittivity of Polymer–Barium Titanate Nanocomposites," *Ind Eng Chem Res*, vol. 60, no. 36, pp. 13265–13271, Sep. 2021, doi: 10.1021/acs.iecr.1c02424.