SUMMARY

Artificial Intelligence (AI) has seen a massive increase in importance over the past years, and with it comes a high power consumption required for operation of the deep learning networks. One way to reduce the power consumption of these systems is to move away from computing on central processing units (CPU), accelerated by graphics processing units (GPU) and look into brain-inspired (neuromorphic) hardware. Current neuromorphic chips already outperform conventional computers in terms of power consumption for certain tasks (pattern recognition), but the large amount of transistors required for these chips in order to emulate one neuron, limits their functionality. As an alternative, and to lower the power consumption even further, research is looking into new materials and novel device structures that go beyond traditional Complementary Metal–Oxide–Semiconductor (CMOS) architectures. In this work, we investigate the creation of self-assembled nanoscale networks of oxide materials, with the intention to develop richer, more complex, and more tunable materials, including memristors. We achieve this through the combination of polymer imprinting and polymer templating to form networks of different metal oxides. (**Chapter 1**)

Chapter 2 describes the experimental methods used throughout this thesis, including both the general concepts, as well as all the concrete experimental parameters of every process. In addition, this chapter also discusses the issues that were encountered during the synthesis process, and includes practical recommendations to those planning to work in this direction.

Chapter 3 reviews the progress and perspective of polymer templating of multifunctional oxide nanostructures and discusses the state-of-the-art of the field at the time when this work was started. This chapter gives an overview on how polymer templating offers an interesting alternative for the fabrication of metal oxide nanostructures compared with other traditional methods for the fabrication of thin film heterostructures, such as lithography, pulsed laser deposition (PLD) and chemical solution deposition (CSD). Templating using sequential infiltration synthesis (SIS) and CSD is discussed, as is the use of multiblock copolymers as a pathway to obtain complex structures. Finally, an outlook on the progression of the field and interesting directions to investigate are provided. This chapter is based on the published paper: Xu, Berg* et al., *J. Appl. Phys.* **128**, 190903 (2020).

In **Chapter 4** we show that highly ordered and interconnected nanoscale networks of functional metal oxides can be fabricated by a combination of polymer imprinting and polymer templating through solution processable methods. We report the fabrication of $BiFeO_3$ (BFO), $SrTiO_3$ (STO), $La_{0.7}Ca_{0.3}MnO_3$ (LCMO), and HfO_2 networks, showing that all of these

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materials are able to follow the self-assembled network morphology dictated by the polymer structure. These networks were extensively characterized using various techniques. Grazing Incidence Small Angle X-ray Scattering (GISAXS) showed that a good imprint quality was obtained on the mm^2 scale for the combined networks, which is challenging given that multiple processing steps were involved during the fabrication. The material stoichiometries were investigated by X-ray Photoemission Spectroscopy (XPS) and the crystal phases were characterized by Grazing Incidence Wide Angle X-ray Scattering (GIWAXS). The electronic functionality was as anticipated: Conducting Atomic Force Microscopy (cAFM) on the $La_{0.7}Ca_{0.3}MnO_3$ networks confirmed the conductive character of this composition; and Piezoresponse Force Microscopy (PFM) of the $BiFeO_3$ network is consistent with the presence of ferroelectric behavior. This chapter is based on the published paper: Berg et al., *ACS Appl. Nano Mater.* **5**, 9, 13349–13360 (2022), and shows that a multitude of materials are in reach for the combined templating and imprinting method and that the materials reported show promise for future applications in adaptable electronics.

Chapter 5 discusses the synthesis and characterization of $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) structures, offering a perspective on the potential use of the LSMO as memristive material. LSMO has a rich phase diagram, with transitions between phases that display different magnetic, structural and electrical transport character, as a function of Sr-doping, and shows a strong coupling between the magnetic and metal-to-insulator transition temperatures (T_C) and T_{MI}, respectively), making it especially interesting for applications. In this chapter, resistive switching in LSMO nanostructures, fabricated through chemical solution deposition and polymer templating, is investigated. For these nanostructures, the dimensions are reduced in both the in-plane and out-of-plane directions and we discuss the effects that this dimensionality reduction has on the transport properties of the networks. From here, we reason that the critical thickness needs to be overcome in order to obtain materials and devices that can show conduction at room temperature. In order to do so, different multilayer structures were fabricated and discussed. Electrical conduction was only observed for the thickest attainable nanostructures, which had a total thickness of ~20 nm. For these samples, switching loops with highly rectifying behavior were observed. Initial conductive AFM (cAFM) studies show resistive switching and non-volatile behavior, although the underlying mechanism remains unclear.

This thesis is concluded by a **General Outlook**. In this short chapter I provide a discussion on the impact of this work and my view on the next steps that should be taken in order to advance the field and create a future for self-assembled networks in neuromorphic applications.