

A digitalization framework enabling development and application of self-assembled materials

Doctoral Dissertation Summary

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July 15, 2025

Block copolymers (BCPs) are macromolecules that can self-organize in distinct morphologies with characteristic dimensions of the order of tens of nanometers. Because of this property, they found broad application in many fields such as nanotechnology, microelectronics, and metrology. However, their fabrication process depends on many parameters interconnected by complex relations, and the manufacturing process is often mainly based on a trial-and-error approach.

In this thesis, a comprehensive standardized database of scanning electron microscope (SEM) images containing ad hoc parameter metadata of lamellar-shaped nanoscale BCP samples is presented. The aim is to provide a complete database that correlates fabrication parameters with morphological characteristics of these nanostructures. The database follows the digital system of unit (Digital SI) principle when reporting the parameter data as metadata and adheres to the Findable, Accessible, Interoperable, and Reusable (FAIR) principles and is also open access for public use. In order to build the database, an algorithm has been developed for the automated image analysis of SEM images. The algorithm analyzes the images and extracts the morphological parameters of the nanostructures to ensure automatization, reproducibility, and standardization of the analysis. The algorithm is generalized to analyze many different types of SEM images acquired with different instruments and taken with various settings. The database and algorithm have been validated through an interlaboratory comparison that involved the characterization of representative samples with the grazing-incidence small-angle X-ray scattering (GISAXS) measurement technique that is traceable to the international system of units.

From an application point of view, the accurate and reliable algorithm developed for analyzing SEM images and extracting the genuine BCP pattern and morphology parameters enabled the study of BCP-based physical

unclonable functions (PUF) for anticounterfeiting applications. The aim is to address the growing counterfeiting problem in today's world. In the thesis, it is discussed how the phase separation given by the self-assembly process of BCPs has proved to be a randomness source that can be exploited for the realization of BCP-based PUF devices. It is detailed how the BCPs are exploited as templates for engraving their artificial fingerprint structure on a wide range of materials and substrates, thus creating robust PUF devices that proved to be resilient to high and low temperature treatment and aging. A robust computer vision technique is proposed for the PUF authentication or identification procedure, enabling their use in real-world scenarios. Hence, it is shown in this work that BCP-based nanoscale PUFs can enable secure authentication and identification of products and objects, overcoming some of the limitations of current anticounterfeiting technologies.

Finally, it is discussed how the developed database can be used to extract value from data through machine learning techniques to enable a material-by-design approach for BCPs and perform rational sample fabrication minimizing the number of experiments required to obtain the desired nanostructures for tailored applications.