Convolutional neural network for high-throughput materials characterization with X-ray photoelectron spectroscopy using the Simulation of Electron Spectra for Surface Analysis code

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X-ray photoelectron spectroscopy (XPS) is a surface sensitive (<10 nm) characterization technique used to investigate material properties including chemical composition, chemical depth distribution and electronic structure [1,2]. Particularly in recent years, XPS has become a reliable and advanced experimental technique in various disciplines of science and engineering resulting in the generation of large spectral data sets. Extracting quantitative information from these data sets has traditionally required trained spectroscopists to perform empirical peak-fitting routines for each individual spectrum. For example, to obtain the atomic concentration of a particular element in a sample, the expert needs to determine the integrated peak area from non-trivial fitting routines based on the inelastic scattering background and zero energy loss line shapes, and normalize it according to relative sensitivity factors [3]. In response to the increasing demand for reliable and instantaneous spectral analysis due to the advancement of XPS at synchrotrons and with modern XPS instruments, we propose an automated quantitative X-ray photoelectron spectrum analysis pipeline by combining the Simulation of Electron Spectra for Surface Analysis (SESSA) software with a convolutional neural network (CNN). SESSA serves as an important tool in the field of XPS either as a database for material parameter retrieval or as a Monte Carlo based simulation software for quantitative interpretation of XPS spectra or Auger electron spectra (AES) for a variety of material structures such as bulk, nanostructures, layered spheres, or core-shell nanoparticles [4,5]. SESSA is based on the partial intensity approach (PIA), which considers the energy loss per number of inelastic collisions during the electron transport, and simulates the electron trajectory using a Monte Carlo simulation with the trajectory reversal approach [5]. The generated electron spectrum consists of the core-shell electron peaks, Auger peaks and a scattering background resulting in relatively realistic spectra. In this work SESSA is used to generate approximately 1.2 million spectra for 7579 inorganic and organic bulk compounds and single elements, illustrated by the histogram overlaid with the periodic table in Figure 1. To increase the variability in the simulated data set and to reflect experimental conditions including thermal and instrumental broadening, we augment our simulated spectra by considering different chemical shifts, different peak widths and different peak shapes (Gauss, Lorentz). These simulated spectra, together with their corresponding chemical abundance labels, were used to train a CNN to classify the stoichiometry of non-crystalline bulk materials (see Figure 2). The aim of the study is to investigate the feasibility of applying deep learning models to high-throughput material characterization within XPS by generating a data set of XPS survey spectra. The current model can already predict the concentration of the most common single elements (Z=5 to 17) studied in XPS (88% accuracy, mean absolute error 0.08) that are constituents of the bulk compound but fails to classify the full stoichiometry of complete complex compounds (20% accuracy) in the test set. In Figure 3 we compare the true and predicted concentration in the test set, illustrating the overall agreement between prediction and ground truth with missing classification especially for atomic numbers (Z > 17) for which we have little data in the data set. An example of a correct classification of an XPS survey spectrum included in the test set is shown in Figure 4. We are currently working on improving the network performance to classify more complex compounds correctly. We are also testing the performance of the network on experimental data and comparing it with classical peak-fitting routines. In the future, we plan to extend our dataset to structured materials, again using our simulation software SESSA for film thickness prediction applied to high-throughput depth profiling which is another key application of XPS in materials analysis [6].

References

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Figures

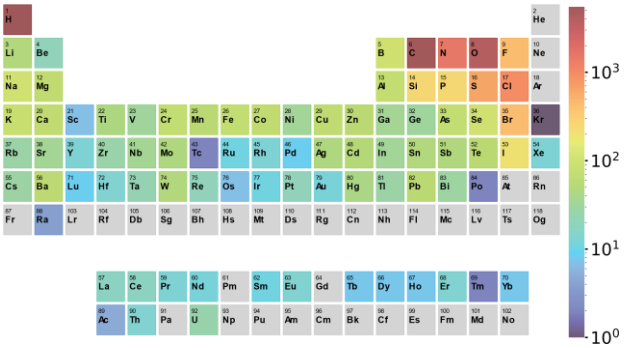

Figure 1. Element frequency in the simulated spectral data set.

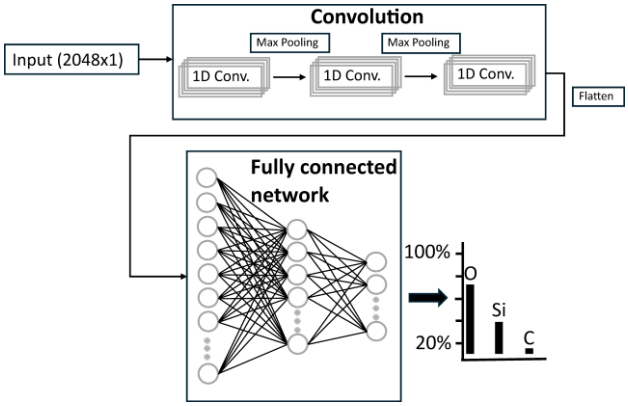

Figure 2. Convolutional Neural Network architecture used to predict the stoichiometry of bulk materials based on XPS survey spectra of input size (2048x1).

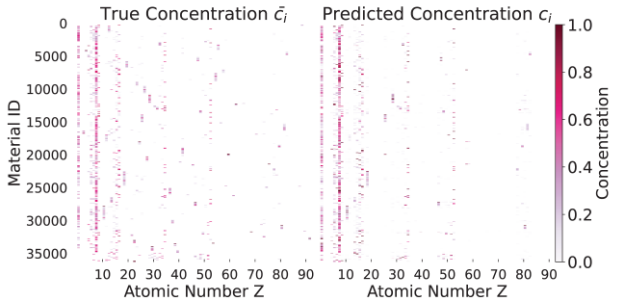

Figure 3. Comparison between actual concentration and predicted concentration of the test set.

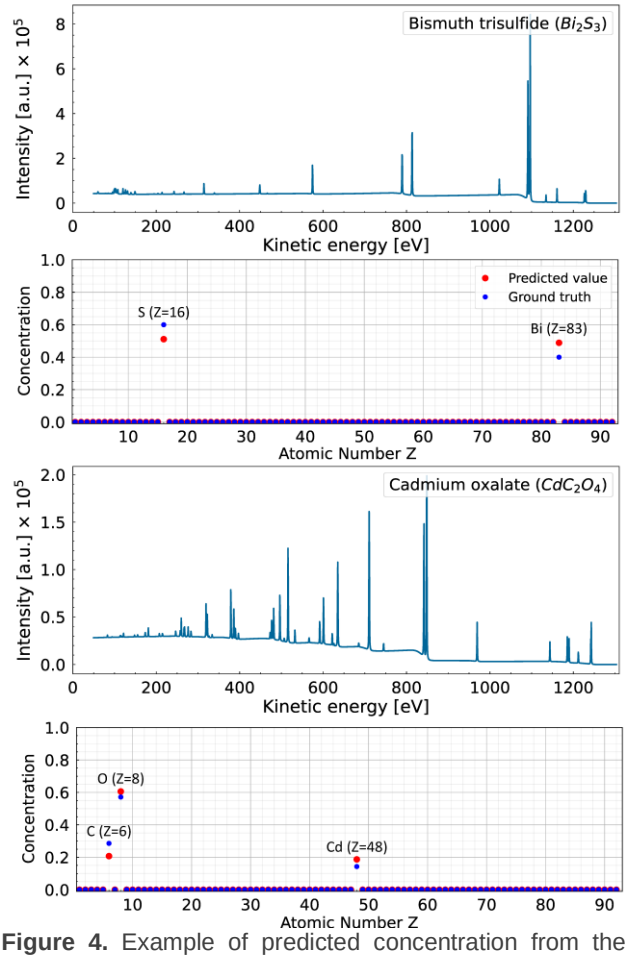

Figure 4. Example of predicted concentration from the XPS survey spectrum compared to the ground truth stoichiometry of the material.