doi:10.1017/S1431927622003269
Microsc. Microanal. 28 (Suppl 1), 2022

Solving Peak Tail Overlaps in Atom Probe Tomography using Convolutional Networks

Martin S. Meier[1], Paul A. J. Bagot[1], Michael P. Moody[1] and Daniel Haley[1]

[1]Department of Materials, University of Oxford, 16 Parks Road, Oxford, OX1 3PH, United Kingdom
* Corresponding author: martin.meier@materials.ox.ac.uk

Atom Probe Tomography (APT) is an emerging, high-resolution microanalysis technique that also achieves high chemical sensitivity, applying time-of-flight (ToF) spectroscopy to ions that evaporate from a sample to measure their mass-to-charge state ratio. Ions contribute to peaks in the resulting ToF or mass-to-charge spectra. The shape of these peaks depends on numerous factors, including the sample as well as experimental, machine and analysis parameters [1]. To date, no model exists that can predict peak shapes across a wide range of experiments.

In the mass-to-charge-state ratio spectra, overlaps of peaks with tails from preceding peaks are a well-known problem in quantitative analysis of APT data (Figure 1). Several methods to resolve these have been applied in the past, e.g. full peak shape fitting [1, 2] or the use of peak heights as a proxy for full peak counts. Fundamentally, peak-tail overlaps are a problem that only can be solved statistically: APT does not provide sufficient information to assign ion identities to individual ions in such overlaps. To decompose relative contributions to peak-tail overlaps, it is necessary to inject information that is not directly contained in the APT spectrum that is to be analysed, e.g. assumptions about certain peaks in a spectrum having a similar shape, or empirical knowledge of typical APT peak shapes [1, 2].

We explore the use of neural networks in order to predict the shape of overlapped peak tails. Our approach is as follows: First, noise-corrected, tail-overlapped peaks from an APT experiment (Figure 2-a) are converted into a spectrum with Gaussian-shaped peaks without overlaps (Figure 2-b), using a convolutional network. This trivially permits us to remove peaks that overlap with peak tails through manual editing of the spectrum (Figure 2-c). Then, the edited Gaussian-peak spectrum is converted back into a spectrum with realistic peak shapes (Figure 2-d). The overlapped peak is now missing, meaning that the predicted tail of the preceding peak (which does no longer overlap) can now be observed in the spectrum, and predicted counts can be measured.

This means the networks need to conduct image translation (a spectrum effectively is a 1-dimensional image), where APT spectra are translated into Gaussian-peak spectra without overlap and vice versa. The necessary training data, which are many APT experiments from which the networks can “learn” typical peak shapes, are obtained from a database of past experiments. One challenge to this approach is the lack of paired training data of real APT experiments and their Gaussian-peak counterparts, preventing the direct, supervised training of networks that directly can predicts counts from spectra. We therefore use cycle consistent generative adversarial networks [3], which can learn the peak shapes unsupervised, sidestepping the paired data problem.

This research has the potential to improve the quantitative capabilities of APT, especially in cases of complex peak-tail overlaps with many different isotopes involved, where the exact peak shape is unknown and peak fitting is not feasible. Our networks learn the structures of APT spectra from past experiments effectively and apply them to predict peak tails in new experiments. This means, we can build upon knowledge existing within a very large amount of historical data from previous experiments in order to resolve peak-tail overlaps in current APT spectra.

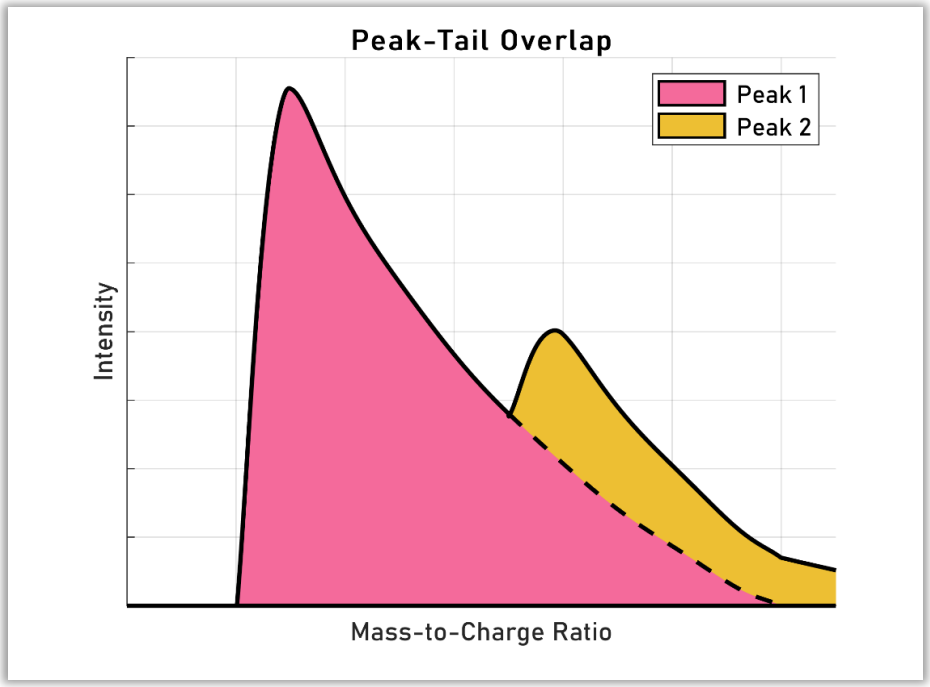

Figure 1: Schematic drawing of a peak-tail overlap in APT. Decomposition of the overlapped areas is necessary in order to obtain accurate counts for both peaks

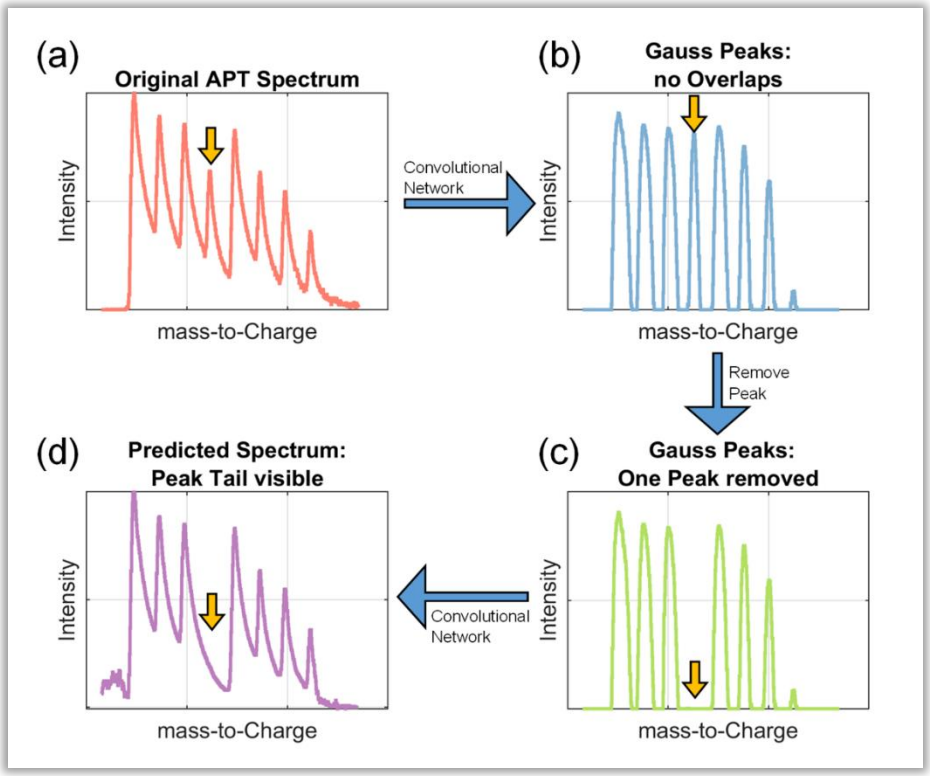

Figure 2: Application of a neural network for removal of a peak from a spectrum in order to predict counts in an overlapped peak tail. An APT spectrum (a) is converted to gauss peaks without overlap (b),

https://doi.org/10.1017/S1431927622003269 Published online by Cambridge University Press

using a convolutional network. The highlighted peak (yellow arrow) is then removed (c), and the so edited spectrum is converted back to an APT peak shape (d). The peak is now missing, and a predicted tail becomes visible.

References

[1] F. Meisenkothen, D. V. Samarov, I. Kalish, and E. B. Steel, "Exploring the accuracy of isotopic analyses in atom probe mass spectrometry," Ultramicroscopy, vol. 216, p. 113018, 2020/09/01/ 2020, doi: https://doi.org/10.1016/j.ultramic.2020.113018.

[2] L. J. S. Johnson, M. Thuvander, K. Stiller, M. Odén, and L. Hultman, "Blind deconvolution of time-of-flight mass spectra from atom probe tomography," Ultramicroscopy, vol. 132, pp. 60-64, 2013/09/01/ 2013, doi: https://doi.org/10.1016/j.ultramic.2013.03.015.

[3] J. Zhu, T. Park, P. Isola, and A. A. Efros, "Unpaired Image-to-Image Translation Using Cycle-Consistent Adversarial Networks," in 2017 IEEE International Conference on Computer Vision (ICCV), 22-29 Oct. 2017 2017, pp. 2242-2251, doi: 10.1109/ICCV.2017.244.

https://doi.org/10.1017/S1431927622003269 Published online by Cambridge University Press