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Methodological development of nanoscopic defects characterization in nuclear materials: contribution of TEM-APT correlative microscopy

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Background

The structural steels used in the primary circuit of nuclear power plants are subjected to irradiation and/or temperature conditions that lead to changes in their mechanical properties. These mechanical properties are directly linked to changes in microstructure due to ageing, particularly on a nanometric scale (segregation, precipitation, clusters of point defects, etc.). To predict these changes, it is necessary to describe the nano-objects present as precisely as possible, i.e. to correlate their chemistry, morphology and crystallography. In the nuclear materials scientist community, two complementary instruments are often used to characterize nano-features: Atom Probe Tomography (APT) and Transmission Electron Microscopy (TEM). APT enables characterization of nm-scale chemical heterogeneities in terms of size, density, composition, and morphology in the 3D of the real space. However, it does not provide a full crystallographic description.
TEM related techniques can provide access to the crystalline structure of the matrix and nano-objects, as well as their orientation relationships. Its wider field of view also allows us to quantify the number density of objects with better accuracy. Chemistry can also be studied in analytical STEM, although the matrix signal can be convoluted with that of the precipitates. Contrary to APT the collected data are only in 2D.

Methods

The aim of TEM-APT correlative microscopy is to characterize the same nano-objects using both techniques for the most complete possible description [1]. Since APT is a destructive techniques, correlative microscopy here consists in analyzing an APT thin needle with TEM first, and then to analyze the same sample by APT. It allows to remove ambiguities about the nature and/or location of nano-objects and thus provide new insights into the mechanisms involved in defect formation.

This work therefore involves developing a methodological approach that makes maximum use of the data collected from each technique (such as MultiVariate Statistical Analysis processing for chemical analysis in STEM) and combination of data accessible by only one of each technique obtained on the same sample. To this end, a thermally aged austeno-ferritic model alloy, in which the ferrite undergoes two phase transformations: spinodal decomposition and the formation of G-phase particles, was studied in an effort to optimize sample preparation for correlative microscopy and data processing. The challenges that have been faced are as follows: improving the contrast of electron microscopy analyses, improving the yields of TEM-APT correlative microscopy analyses, understanding the artefacts associated with the physics of field evaporation and 3D reconstruction for APT, and optimization of data processing of EDS through statistical methods (using Principal Components Analysis in particular) in order to improve EDS quantification and correlation with atom probe data.

Results

The aim of the first part of the study is to extract more quantitative data from TEM analysis (imaging and elemental analysis). Contrasts are difficult to interpret in TEM experiments because of the shape of APT samples, the amorphous layer induced by FIB preparation, the presence of oxide layers hindering diffraction patterns and the contribution of the background (i.e. vacuum) in images. In order to reduce the amorphous layer, the addition of a cleaning step has been investigated with the use of an Ar-beam polishing at low-voltage with a Precision Ion Polishing System II (PIPS II) [2]. In addition to a systematic plasma cleaning before and after every S/TEM analysis, this step would also permit to address the yielding of tips that have been investigated in EM by removing the high-field carbon contamination and the oxide layers simultaneously. Quantification in Analytical TEM is not straightforward when trying to isolate the signal from the precipitates from that of the matrix. Compositions of the α/α' domains as well as G-phase precipitates measured thanks to APT have been used to improve the data processing of EDS data. On one hand, EDS spectrum imaging supported by PCA denoising as well as sample preparation improvement, helped to optimize the detection of nano-objects. On the other hand, thanks to correlative microscopy and PCA, we were able to extract their composition from EDS maps.

The second part of the study is to use TEM images as additional information to obtain reliable 3D reconstructions with atom probe data, even without the presence of crystallographic poles.

Through the comparison of different 3D reconstruction methods that exist on an experimental dataset from TEM-APT correlative microscopy: static and dynamic voltage-based reconstructions [3], shank angle or tip profile, we suggest a methodology to reconstruct a volume properly. This is done by adjusting the spatial distribution of the G-phase particles and their shape. This correlation also enabled to highlight distortion artefacts observed on APT volumes.

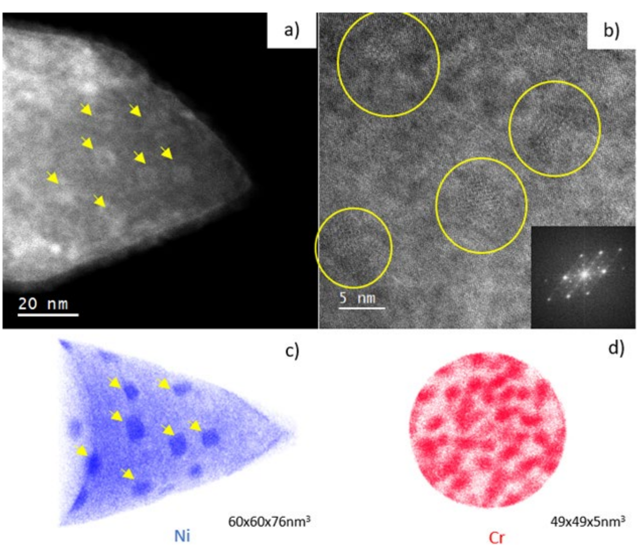
Conclusion and Perspectives

This work aims to present a methodology for extracting maximum information from samples thanks to correlative TEM/APT microscopy. Leveraging the strengths of each instrument, we addressed the limitations of the other, enabling successful characterization of particle shapes, sizes, crystallography and chemical composition. Efforts have been dedicated to improve the sample preparation procedure with low voltage ion beams, allowing improved image contrast and better identification of objects and therefore facilitating quantitative analysis. The optimization of the data processing of EDS spectrum, assisted by APT data, has been undertaken. PCA has demonstrated its effectiveness in reducing the background noise and thus improving the detection of poor signals (e.g. low counts experiments, from nano-particles embedded in the matrix).

Other future developments currently under investigation include:

- Exploring alternative methods to enhance contrast in diffraction-based EM imaging, such as through flash-polishing techniques. [4]
- Expanding application of the methodology to analyze more complex materials that contain a wider range of nano-objects, such as irradiated steels.

Graphic:



Keywords:

TEM-APT correlative microscopy, nuclear metallurgy

Reference:

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