Nanoscale mapping of point defect concentrations via temperature, irradiation, and corrosion with 4D-STEM

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Vacancies are missing atoms in a crystalline material, and occur both at equilibrium (varying with temperature) and out of equilibrium such as when crystalline materials are damaged with radiation or corrosion. While we know of their importance, particularly regarding diffusive mechanisms, it is not straightforward to experimentally measure their concentration or to visualize them directly. Traditionally, measurements such as positron annihilation spectroscopy and to some degree X-ray diffraction can measure average concentrations, but generally lack the ability to visualize or quantify a heterogenous concentration of vacancies that can occur at the level of individual defects and microstructural features. Here, we present a method to map vacancy concentrations and their distribution with 1 nm resolution. Our method utilizes a Au thin film as a model to demonstrate the method via four-dimensional scanning transmission electron microscopy (4D-STEM) by correlating the differences between changes in lattice parameter and the volumetric thermal expansion during in situ heating experiments. The vacancy mapping methodology is also applied to non-equilibrium defects accumulated in pure Al via knock-on electron beam irradiation. Further, we measure nano-scale changes in structure and composition of NiCr exposed to molten salt corrosion via simultaneous STEM-EDS and 4D-STEM mapping. This multimodal approach highlights the significance of chemistry and structure on isolated corrosion pore formation and the uniquely altered morphology that results from non-steady-state nuclear reactor environments.

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