

Review of actinide core-level photoemission F

Cite as: J. Vac. Sci. Technol. A **42**, 050802 (2024); doi: [10.1116/6.0003534](https://doi.org/10.1116/6.0003534)

Submitted: 12 February 2024 · Accepted: 5 June 2024 ·

Published Online: 23 July 2024



Alaina Thompson,^{1,2} William Limestall,^{1,3} Art Nelson,⁴ Daniel T. Olive,² and Jeff Terry^{1,5,a)}

AFFILIATIONS

¹Department of Physics, Illinois Institute of Technology, 3101 South Dearborn Street, Chicago, Illinois 60616

²Materials Science and Technology Division, Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, New Mexico 87545

³Chemical & Fuel Cycle Division, Argonne National Laboratory, 9700 S. Cass Avenue, Lemont, Illinois 60439

⁴Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, California 94551

⁵Department of Mechanical, Materials, and Aerospace Engineering, Illinois Institute of Technology, 3101 South Dearborn Street, Chicago, Illinois 60616

Note: This paper is part of the Special Topic Collection: Reproducibility Challenges and Solutions II with a Focus on Surface and Interface Analysis.

^{a)}Electronic mail: terryj@iit.edu

ABSTRACT

Photoelectron spectroscopy allows for the investigation of the electronic structure and chemical bonding of actinide elements and their compounds, providing insights into oxidation states, chemical environments, and electronic configurations. This knowledge can aid in comprehending reactivity, stability, and other properties of actinide materials, which is essential for ensuring safe handling, storage, and disposal in nuclear applications. We have reviewed a number of results in actinide core-level photoemission studies, with a particular focus on x-ray photoemission spectroscopy (XPS) techniques. Actinides, due to their inherent radioactivity, have not been as well studied with XPS as have other segments of the periodic table. Given the inherent safety concerns, equipment requirements, and short isotopic lifetimes associated with actinide research, we outline the strategies and precautions necessary for conducting successful and safe XPS experiments on these elements. Core-level photoemission can be a powerful proven tool for investigating the electronic structure, chemical bonding behaviors, and physical properties of actinides, providing valuable insights into an incredibly complex behavior of these systems. We highlight key findings from recent studies that demonstrate the potential of core-level photoemission in uncovering the unique properties of actinides and their compounds. Finally, we identify current knowledge gaps and future research directions that could enhance our understanding of actinide chemistry and physics.

Published under an exclusive license by the AVS. <https://doi.org/10.1116/6.0003534>

I. BRIEF INTRODUCTION TO CORE-LEVEL XPS

X-ray photoemission spectroscopy (XPS), also known as electron spectroscopy for chemical analysis (ESCA) and photoelectron spectroscopy (PES), has the ability to provide information on oxidation state, chemical environment, and bonding characteristics of uranium surfaces.¹ Photoemission spectroscopy's theoretical basis was laid over a century ago, with Albert Einstein's groundbreaking paper "On a Heuristic Point of View Concerning the Production and Transformation of Light," for which he won the 1921 Nobel prize. Researchers' ability to perform photoemission measurements on most materials did not come of age until the 1960s. It was at this time that the equipment in both x-ray sources and data collection systems, as well as analysis capabilities, matured enough to

become an emerging technology. In tandem, development of new materials including semiconductor compounds were also gaining traction; moreover, complex interactions were becoming more understandable using newly developed and robust solid-state theories of quantum mechanics.² Core-level photoemission studies give insights into the chemical bonding, electronic structure, and physical properties of materials through investigations of electronic states associated with the core electrons of atoms in solids. It is generalized that core electrons are less involved in chemical bonding.³

The 1976 "International conference on the electronic structures on the actinides" showed that XPS was an incredibly powerful technique for determining not just how the 5f electrons behave in

05 September 2024 20:11:42