XAS Studies On Nuclear Fuels: From Simulated to Spent Fuels Characterizations

V. Klosek[1], M. Rochedy[1], W. Tomczak[1], C. Le Gall[1], S. Gobert[1], A. De Azevedo[1], F. Audubert[1], O. Proux[2], I. Kieffer[2], J.L. Hazeman[2], M.O.J.Y Hunault[3], D. Menut[3], P.L. Solari[3]

1 CEA, DES, IRESNE, DEC, Cadarache, 13108 Saint-Paul-lez-Durance, France
2 BM16/FAME-UHD, ESRF, 71 avenue des Martyrs, Grenoble F-38000, France
3 Synchrotron SOLEIL, L'Orme des Merisiers, Saint Aubin, BP 48, 91192 Gif-sur-Yvette

ABSTRACT

Fission of U and Pu during fuel irradiation in nuclear reactors leads to formation of more than 60 different chemical elements, in small or even very small quantities (< or << at.%). Determining the chemical and microstructural roles of the fission products (FPs) in the evolution of nuclear fuel physical properties is of crucial importance to develop reliable long-term predictions of the fuel behavior during reactor operation under normal and accidental conditions as well as for storage and recycling of spent fuel.

X-ray absorption spectroscopy (XAS) is the element-specific technique of choice to determine the speciation of an element, even very diluted, in potentially complex samples. However, the accurate determination of nuclear fuel chemistry is a very challenging task. First, fission products in UO_2 create a complex multi-component system, in which a given element can form various phases which may be different depending on the location within the pellet. Secondly, spent nuclear fuel is highly radioactive and requires specific radiological safety devices and protocols to be handled and studied. In some cases, where specific fission products are to be characterized under particular conditions, irradiated fuels can be replaced by model, low activity materials, such as SIMFUEL (UO_2 doped with stable isotopes of fission products) or ion-implanted UO_2 pellets.

Numerous XAS studies have been carried out at SOLEIL and ESRF on such model samples to characterize the chemical states and/or behaviors of various FPs (Mo, Xe, Kr, Cs, I, Ba, Zr, etc) in UO_2 as a function of concentrations, temperature and oxygen potential. These "separate effects" experiments aimed at, e.g., feeding thermodynamical databases with experimental data to improve modelling [1], or improving the understanding of the behavior of FPs during a severe accident [2]. They also help developing innovative advanced fuels.

Recently a massive sample of UO_2 spent nuclear fuel could be characterized for the first time by HERFD-XANES on the MARS beamline to determine the chemical states of Mo and Cs [3]. This first experiment opens up many prospects for future XAS characterizations of spent fuels.

An overview of all these XAS studies on model or spent fuels will be given. The technical and scientific challenges, as well as the required technical and methodological developments will be exposed.

REFERENCES

[1] M. Rochedy et al. J. Nucl. Matter, 581, 154450 (2023).
[2] C. Le Gall et al. J. Solid State Chem., 319, 123792 (2023).
[3] V. Klosek et al. J. Nucl. Matter, 586, 154660 (2023).