Proceedings of the INMM & ESARDA Joint Virtual Annual Meeting
August 23-26 & August 30-September 1, 2021

ASSESSING THE CAPABILITIES OF DIFFERENT HKED SPECTRUM PROCESSING ALGORITHMS

G. Toma[i], K. Casteleyn, A. Le Terrier, M. Toma, J. Zsigrai and E. Zuleger
European Commission, Joint Research Centre (JRC), Karlsruhe, Germany

ABSTRACT

The Hybrid K-Edge/K-XRF Densitometry (HKED), is the leading radiometric technique for the assay of uranium and plutonium in nuclear safeguards samples and for process control in nuclear fuel reprocessing. It is based on X-ray transmission and X-ray fluorescence spectrometry. The Joint Research Centre (JRC) has organized an exercise to assess the capabilities of HKED spectrum processing algorithms and to identify areas for improvement. Within the exercise, the JRC distributed spectra recorded by HKED systems operated by the JRC to the international community of HKED users. The first set of spectra was distributed in May 2019 and another one in March 2020. These sets included spectra of well-characterized nuclear fuel reprocessing input solutions, and of dissolved uranium and MOX samples. The results of the spectrum analyses were better with the second set than with the first set, indicating likely improvements of the algorithms. All algorithms performed excellently with spectra of dissolved uranium and MOX pellets and not as good with reprocessing input solutions. The reasons for this behaviour need to be better understood and addressed in the algorithms. A third round of joint analysis, with a new set of spectra, is in preparation.

INTRODUCTION

Developed in the 1980s at KfK Karlsruhe, Germany, the Hybrid K-Edge/K-XRF Densitometry (HKED) technique is used for accurate and timely measurement of U and Pu element concentrations in liquid samples (for example solutions of reprocessed spent fuel). Compared to other analytical methods, HKED is able to process a larger number of samples in shorter time. As a result, it has become the main analytical technique applied in nuclear fuel reprocessing plants for nuclear safeguards and for process control.

The first HKED system for safeguards was deployed at the La Hague reprocessing plant, in France in 1989 by the Euratom Safeguards Directorate (ESD). Today the technique is employed worldwide. Despite the geographically wide adoption, there are not many HKED systems and the community of users is small. This is mostly because the hosting locations are not numerous themselves: fuel reprocessing plants and nuclear research laboratories.

The HKED systems have evolved over the years rather independently from one another. The basic design template was adapted to local conditions of operation. The hardware and software

upgrades that are underway are also unfolding rather independently from one site to the other. The JRC has identified an opportunity to bring together the users of HKED systems worldwide in order to facilitate contact and communication in particular on the topics of performance and upgrade. In collaboration with other members of the HKED community, JRC has organized workshops dedicated to the HKED [1].

THE HKED TECHNIQUE

The HKED systems use two high-purity germanium (HPGe) detectors commonly called the KED and the XRF detector [2]. The dissolved U/Pu samples are placed in an X-ray beam and the two detectors are used to measure different components of the X-ray radiation (see Figure 1). The KED detector is used to measure the transmitted intensity (densitometry measurement), while the XRF is used to measure the emitted characteristic U/Pu X-rays. The HKED can be used to assess the U and Pu concentration in solutions under 3 different scenarios:

I. The pure KED mode is used for densitometry measurements. These are quantitative measurements of "optical" density in a medium. The measured effect is the decrease in the intensity of X-ray due to absorption at the specific K-edge energy of the dissolved substance (U or Pu). This mode is used to measure concentrations in concentrated pure Pu or U samples ([U], [Pu]>50 g/L) or in mixtures with a dominant element (for the dominant element).

II. The pure XRF mode is used for recording the emission of characteristic "secondary" (or fluorescent) X-rays from a material that has been excited by being bombarded with high-energy X-rays or gamma rays. The XRF mode is applied to measure very low U, Pu concentrations (below 50 g/L) by absolute XRF or to measure atom ratios in U/Pu mixtures.

III. In the hybrid mode of operation (HKED mode), by combining KED and XRF measurements for a mixture with a dominant element, we can infer the concentration of the minor element.

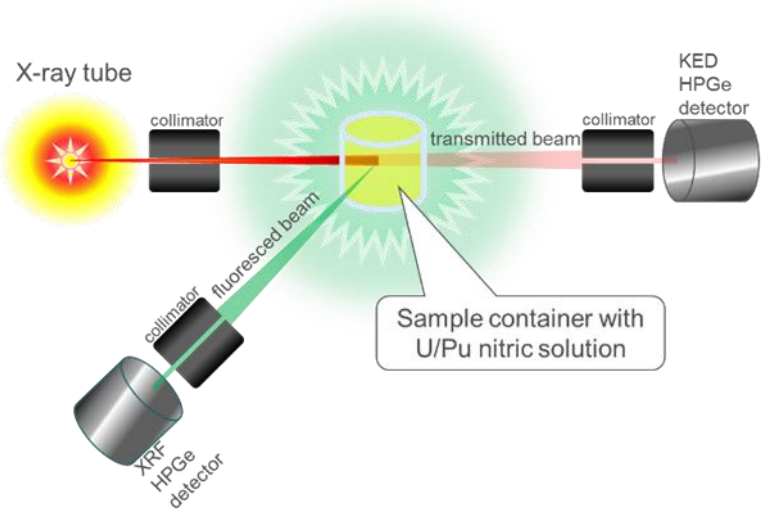

Figure 1. Simplified representation of the HKED detector system

THE EXERCISE

The Participants

The JRC has initiated thematic workshops dedicated to the HKED technique. These workshops are planned to take place on a regular basis and their main goal is to bring together the community of HKED users in order to foster communication (as a short-term objective) and collaboration for future developments of the method (as a long-term objective). Two workshops have been organized so far (in 2018 and 2020) and a third is already envisaged.

In correlation with these workshops, the operators of HKED systems were given the opportunity to participate in a data analysis exercise in order to assess the capabilities of their spectrum processing algorithms. Laboratories of the CEA[ii], IAEA[iii], IPPE[iv], JRC and ORNL[v] have participated to the exercise.

The Data

For this exercise, the JRC has distributed two sets of HKED spectra of well-characterized input solutions from nuclear fuel reprocessing and of dissolved pure uranium and mixed oxide fuel (MOX) samples. One set was distributed in May 2019 and another one in March 2020. For each data set, three samples of each type were selected. The U concentration range was ≈107 g/L to ≈225 g/L and for those samples containing Pu, the U/Pu ratio was in the range ≈10 to ≈125 (see Table 1 for set no. 1 and Table 2 for set no. 2). The concentration values in Table 1 and Table 2 are derived from mass spectrometric measurements. For each sample, JRC communicated also the following additional data: solution density, sample temperature during measurements, X-ray path length through solution, U enrichment, Pu molar mass and the intensity of the current used for X-ray production. JRC provided also reference matrix spectra (of pure acid solutions). A measurement date and a reference date were provided for the samples containing Pu, to account for Pu decay.

Table 1. Overview of samples used to build the first data set. The values of [U] and [Pu] and their respective relative uncertainties δU and δPu are given by mass-spectrometric measurements.

Sample ID (type)	[U] [g/L]	δU [%]	[Pu] [g/L]	δPu [%]	U/Pu mass ratio
Input 1	224.97	0.15	1.8045	0.15	124.67
Input 2	160.50	0.15	1.2873	0.15	124.67
Input 3	109.15	0.15	0.8755	0.15	124.68
MOX 1	141.01	0.07	14.9009	0.13	9.46
MOX 2	153.37	0.07	10.5660	0.13	14.52
MOX 3	153.42	0.07	10.4682	0.13	14.66
Pure U 1	169.27	0.07	NA	NA	NA
Pure U 2	176.79	0.07	NA	NA	NA
Pure U 3	178.46	0.07	NA	NA	NA

Table 2. Overview of samples used to build the second data set. The values of [U] and [Pu] and their respective relative uncertainties δU and δPu are given by mass-spectrometric measurements.

Sample ID (type)	[U] [g/L]	δU [%]	[Pu] [g/L]	δPu [%]	U/Pu mass ratio
Input 1	220.82	0.15	2.1004	0.15	105.13
Input 2	160.55	0.15	1.5272	0.15	105.13
Input 3	106.59	0.15	1.0139	0.15	105.13
MOX 1	145.13	0.07	15.2459	0.13	9.52
MOX 2	149.39	0.07	10.2421	0.13	14.59
MOX 3	145.97	0.07	15.3441	0.13	9.51
Pure U 1	176.26	0.07	NA	NA	NA
Pure U 2	175.66	0.07	NA	NA	NA
Pure U 3	168.65	0.07	NA	NA	NA

The reprocessing input solutions were analysed by JRC using a HKED system installed in the EURATOM safeguards laboratory in La Hague, France, operated by the JRC, while the MOX and uranium samples were analysed using a HKED setup in JRC-Karlsruhe.

RESULTS AND DISCUSSION

The participants analysed the spectra with their own software tools and independently from each other. JRC collected afterwards the results. One participating laboratory has contributed to this exercise with two different analysis algorithms. Hence, even though the legend lists 6 algorithms, there were in fact always only 5 participating laboratories to the exercise.

In Figure 2 we show the performance of different algorithms when applied to the first data set (top) and to the second data set (bottom). The plots show the relative differences between the values obtained by participants and the reference value obtained by mass spectrometric measurement. The sample type (input, MOX or pure U) is indicated for each plot under each set of points. In order to facilitate a visual comparison between the two sets of results, we are using the same vertical scale for both sets. The same marker and line type is used for a given algorithm throughout all plots. The lines are eye guides to help the reader follow the performance of a given algorithm for the various sample types.

The results of the spectra analyses were better with the second set than with the first set. This is due in part to the fact that the participants have used the results from the first set in order to fine-tune their algorithms and compensate for unknown variables (such as different geometry effects between JRC and their experimental setups). Of course this does not exclude the possibility of other improvements that were added to the HKED algorithms in the period between the two exercises. All algorithms performed well with spectra of dissolved uranium and MOX pellets. For the case of reprocessing input solutions the performance was not as good, in particular for the first set. The reasons for this behaviour need to be investigated and better understood.

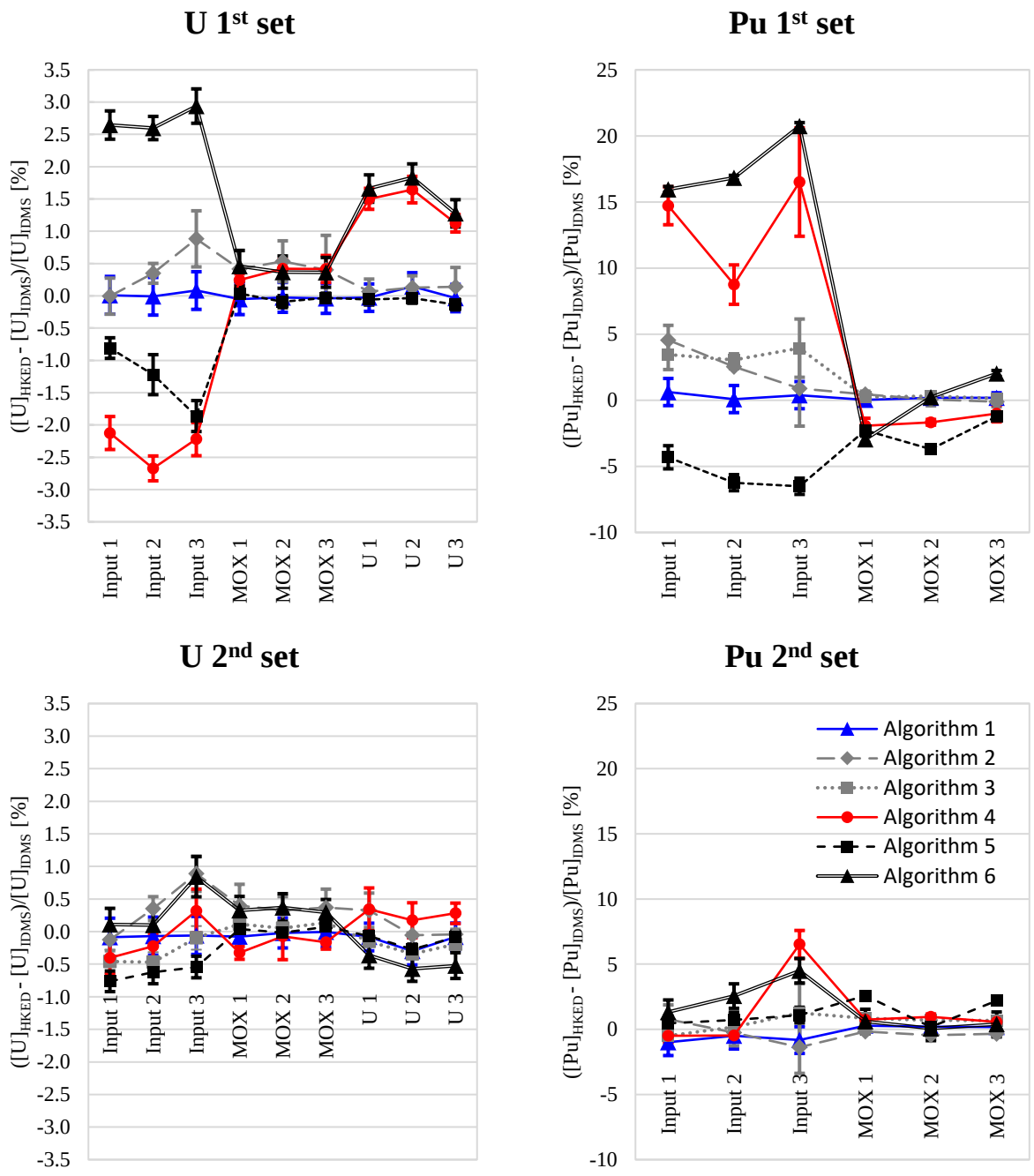

Figure 2. Relative difference between HKED result and reference IDMS measurement for each participant for U (left) and Pu (right).

CONCLUSIONS AND FUTURE PLANS

The HKED workshops achieved their goal of bringing together the community of HKED users. In addition, the results of the joint exercise suggest that the international collaboration on HKED spurred the further development of HKED algorithms. These developments have created the premise for a third round of the joint exercise, with a new set of spectra.

ACKNOWLEDGMENTS

This paper was possible through the contributions from the participating laboratories of CEA, IAEA, IPPE and ORNL. We are grateful to Eric Esbelin and Cédric Rivier (CEA), Andrey Bosko, Georges Duhamel and Dante Nakazawa (IAEA), Sergei Bogdanov and Dmitrii Volnistov (IPPE) and Robert McElroy Jr. (ORNL) for the insightful discussions during the HKED workshops and for their participation in this exercise.

REFERENCES

[1] Zsigrai, J., Le Terrier, A., Zuleger, E., Luetzenkirchen, K., Toma, M., Morel, S., Bosko, A., & Duhamel, G. (2019). Shaping Together the Future of the Hybrid K-Edge / K-XRF Technique. (IAEA-CN--267). International Atomic Energy Agency (IAEA).

[2] Ottmar, H., & Eberle, H. (1991). Hybrid K-Edge / K-XRF Densitometer: Principles-Design-Performance. Kernforschungszentrum Karlsruhe, KfK 4590, ISSN 0303-4003.

---

[i] Presenter
[ii] Le Commissariat à l'énergie atomique et aux énergies alternatives (CEA), France
[iii] The International Atomic Energy Agency
[iv] Joint Stock Company "State Scientific Centre of the Russian Federation – Institute for Physics and Power Engineering named after A.I. Leypunsky", Russian Federation
[v] Oak Ridge National Laboratory, USA