

TABLE 8 - COMPUCEA

Method/measurement system (including software): (COMPUCEA)**Application:** On-Site Uranium Elemental Assay and ^{235}U Enrichment Determination

Short Description of the method/measurement system and typical measurement conditions (shielding, measurement time, etc.)
The COMPUCEA (COMbined Procedure for Uranium Concentration and Enrichment Assay) is a transportable analytical measurement system that combines L-edge transmission and γ-ray spectrometry methods for high-accuracy (bias defect) on-site uranium concentration and enrichment assays of various uranium bearing materials (pellets, powders, scrap, liquids) and compounds (UO_x, ADU, AUC, UF_6, UNH, UO_2F_2). A solid sample with a typical mass of 5 g is quantitatively transferred into a uranyl nitrate solution. A multistep procedure, involving dissolution in 8 M nitric acid and subsequent dilution to target 3 M acidity and target 190 gU/l concentration is employed. In the case of a concentrated liquid sample, a dilution step may be required, to bring the uranium concentration inside the L-edge linearity interval (<200 gU/l). The obtained solution is characterized for its density and temperature. Aliquots taken from the prepared sample solution are analyzed by L-edge absorption densitometry for uranium concentration assay and passive gamma-ray counting for enrichment determination. The L-edge densitometer is based on a high-resolution Peltier-cooled Si drift detector (e.g. 10 mm² x 0.45 mm SDD), a miniature 30 -50 kV X-ray generator, and a 2 mm pathlength flow-through quartz cell fixed inside a Ti shield. The gamma-ray spectrometric part consists of two $\varnothing 2.5'' \times 0.5''$ $\text{LaBr}_3(\text{Ce})$ detectors, arranged inside tungsten shields. A 10 mL aliquot of solution to be analyzed is loaded into a polystyrene container and counted in a fixed geometry. Three replicates of each measurement type are performed resulting in 3 x 1000 s transmission and 3 x 2000 s gamma-ray spectra. The sample preparation and radiometric measurement data are combined, to evaluate the uranium mass fraction (uranium factor) and the ^{235}U mass fraction (enrichment). In the case of neutron poison additives presence, the measured enrichment is corrected based on the effective Gd_2O_3 content, which is determined from the extrapolated and non-extrapolated fits of the L-edge spectrum. The method is field-calibrated using certified reference material (e.g. CRM-125A) and well-characterized working standards in the form of sintered UO_2 pellets. Daily quality check (QC) measurements are performed to verify the stability of the calibrations in the course of a measurement campaign.
Factors affecting the method performance
The uncertainties shown in Tables 8a and 8b are typical values for cases that are not affected by the factors listed in this section. The uncertainties are normally higher for clean powder samples than for sintered UO_2 pellets, due to moisture uptake, inhomogeneity, and other reasons. This is true for any analytical technique that is accurate enough to see the influence of these factors. However, COMPUCEA is the first technique for which this is explicitly indicated in the ITV tables. Moreover, measurements of low concentrated materials that significantly deviate from the optimal concentration (190 gU/l) would suffer from the low counting statistics in both transmission and gamma spectrometric channels. The absorption measurements of low concentrated solutions may benefit from the use of a longer (e.g. 10 mm) pathlength transmission cell. A significant deviation of the solution acidity from the reference value (3M) will bias the results of the concentration measurements. Large quantities of undissolved residue material (e.g. in the case of dirty scrap) will increase the uncertainty of the uranium factor determination. In the case of materials containing burnable additive (Gd, Er, etc.), the measured enrichment can be negatively biased by up to 0.5%, if respective correction is not applied. If the correction is applied, it tends

to increase variability of ^{235}U enrichment results. Thus, a somewhat larger random uncertainty for ^{235}U enrichment is to be expected in this case. Note that materials containing burnable additives are not considered in Table 8b. Analysis of compounds that are not readily dissolved in nitric acid (e.g. UF_4 , uranium silicide) would require development of dedicated sample preparation procedures.

Source of bottom-up value (only for prospective ITV not based on historical data)

ITVs for UNH solutions (150-400 gU/l): ITVs were derived from the ITVs for solids by increasing the systematic component to reflect the effect of dilution (from 400 gU/l to 190 gU/l) and increasing the random component to reflect the reduced counting statistics at <190 gU/l. The random and systematic uncertainty components for the enrichment determination were adjusted accordingly to account for the increased uncertainty components of respective concentration measurement.

ITVs for UF_6 powder: ITVs were derived from the combination of the UF_6 validation study³ and historical data.

Method references

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[2] A. Berlizov, A. Schachinger, K. Roetsch, N. Erdmann, H. Schorle, M. Vargas, J. Zsigrai, A. Kulko, M. Keselica, F. Caillou, V. Unsal, A. Walczak-Typke, Feedback from Operational Experience of on-site Bias Defect Analysis with COMPUCEA, Journal of Radioanalytical and Nuclear Chemistry, Vol. 307(4) (2016) 1901-1909.

[3] M. Toma, H. Schorlé, M. Vargas Zúñiga, J. Zsigrai, A. Berlizov, A. Schachinger, S. Richter, J. Truyens; Validation of the preparation method and analysis of uranium hexafluoride samples with the COMPUCEA system, JRC Karlsruhe, 2017, PUBSY No. 105447

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