Four decades of progress in PWRs surveillance program dosimetry measurements at the MADERE platform

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Abstract. Since 1981, the French Alternative Energies and Atomic Energy Commission has been measuring the dosimeters of the surveillance capsules of the French pressurized water reactors and assessing their neutron fluence. This represents more than 250 surveillance PWR capsules, i.e. 4 thousands dosimeters. Started in Grenoble, this work continued in Cadarache with the creation of the "Measurement Applied to DosimEtry in Reactors" (MADERE) platform within the Dosimetry, Sensors and Instrumentation Laboratory (LDCI). Since 1998, all surveillance dosimeters measurements (i.e. Al-Co, ^{238}U, ^{237}Np, Ni, Cu, Fe and Nb) have been performed at MADERE under the “Cofrac Essais” accreditation according to the ISO/IEC 17025 international standard. The evolution over time of the 1σ measurement uncertainty of the activity of each is presented in this paper. Since 2015, they are [±1.3% ; ±2.9%] for ^{238}U with Ni encapsulation, [±0.8% ; ±2.1%] for ^{238}U with Ti encapsulation, [±1.2% ; ±4.9%] for ^{238}U with stainless steel encapsulation, ±1.3% for ^{237}Np dosimeters, ±1.2% for Al-Co, [±1.2% ; ±1.4%] for nickel, copper and iron, [±3.3%; ±3.9%] for niobium if unbroken after irradiation.

1 French PWRs and dosimetry in surveillance capsules

France is the 2nd country in the world in terms of number of civil PWRs in operation and total cumulative electricity production: 3 shut down, 56 in operation, 1 under construction and 6 more planned [1]. The old, current and future PWRs have been, are and will be operated by the main French public electricity producer: EDF.

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For French pressurized vessels, it is mandatory to set up a monitoring program for the irradiation of PWR vessels, called irradiation surveillance program ("ISP" in short, "PSI" in French) [2]. To determine the embrittlement of the vessel, it was decided to equip each PWR with several irradiation capsules in the vessel before commissioning. These capsules contain representative specimens of the different materials making up the vessel and families of dosimeters becoming radioactive at different neutron energies. Since the start of the French PWRs, ISP nuclear analyses have been carried out at the CEA, currently at the Dosimetry, Sensors and Instrumentation Laboratory (LDCI) in Cadarache. The LDCI currently measures the activities of all dosimeters thanks to its ISO/IEC 17025 accredited MADERE platform, then uses a methodology to provide the nuclear quantity of interest for each specimen: the fluence of neutrons with energy greater than 1 MeV [3]. The mechanical properties of the specimen are established by EDF in its facilities in Chinon nuclear power plant, also under ISO/IEC 17025 accreditation. Each ISO/IEC 17025 accreditation is supervised and issued by the French accreditation committee: Cofrac. Thanks to the measurement of mechanical property versus the fluence, EDF is able to assess irradiation embrittlement. Thus, the diversity of dosimeters and the reduction of their measurement uncertainties reinforce the demonstration of safety to the French Nuclear Safety Authority.

The types of dosimeters implemented in the ISP capsules vary over the 5 main phases of construction of French PWRs but the irradiation capsules follow a similar pattern: ~1 m-high, ~3 cm thick and ~3 cm wide capsule placed laterally between the core and the vessel at the maximum neutron flux altitude. The first phase of standardized PWRs concerns three-loop reactors of approximately 900 MW_e power (Westinghouse design) divided into two slightly different designs. The first six 900 MW_e PWRs, called "CP0", were equipped with a bare aluminium-cobalt (Al-Co) alloy dosimeter for its ^{59}Co (n, g) ^{60}Co thermal and epi-thermal reaction, bare pure nickel dosimeter for its ^{58}Ni (n, p) ^{58}Co fast reaction and bare pure copper for its ^{63}Cu (n, a) ^{60}Co fast reaction. Each set of dosimeters is tripled to cover the capsule axially: at "high" (H), "medium" (M) and "low" (L) altitudes. To cover the energy range close to 1 MeV, a dedicated "fissile" altitude (F) is installed near level M. It contains uranium (highly enriched in ^{238}U) and neptunium (specifically enriched in ^{237}Np) dosimeters. Each is made of an oxide encapsulated in a nickel case and covered with a layer of cadmium oxide powder tamped by hand (to minimize parasitic thermal reactions while avoiding the risk of melting of cadmium in its metallic form). For subsequent 900 MW_e PWRs (a modified three-loop design called "CPY"), the capsule design changed slightly. At all altitudes H, M and L are added an Al-Co dosimeter shielded with a metallic layer of cadmium (to reduce thermal neutron contribution to its main reaction) and a bare iron dosimeter for its ^{54}Fe (n, p) ^{54}Mn fast reaction. In addition, the neptunium used for the CPY capsules has been selected to be particularly pure in ^{237}Np in order to avoid parasitic fission of plutonium. Finally, the encapsulation material for uranium dosimeters was changed from nickel to stainless steel. The objective was, during the post-irradiation dismantling of the capsules, to make it easier to distinguish the neptunium and uranium dosimeters in the hot cell using a simple magnet.

The second construction phase concerns twenty 1300 MW_e four-loop PWRs divided into a first contract named "P4" and a second named "P'4". For P4 and the first P'4 reactor, the design of the capsules is identical to that of the CPY. EDF in fact accepted LDCI's recommendations concerning the improvement of the capsules shortly after the construction start of the P'4 reactors. First, stainless steel was replaced by nickel for encapsulation of uranium dosimeters in order to reduce the single-escape peak phenomenon due to ^{60}Co (from traces of ^{59}Co) from the 2nd to the 4th P'4 reactor. Then, nickel was replaced by titanium for uranium and neptunium dosimeters encapsulation. Second, solid cases of boron nitride were used from the fifth P'4 reactor to filter thermal neutrons for uranium and neptunium dosimeters instead of the hand-tamped cadmium oxide powder. Third, a niobium dosimeter was added from the fifth P’4 reactor at each H, M, L altitude per capsule to better assess the neutron fluence unfolding.

The third phase of the deployment of French PWRs is the construction of four 1450 MW_e reactors called “N4”. The only change in dosimetry compared to P’4 is the systematic use of dosimeters supplied by the Institute for Reference Materials and Measurements, in particular extremely pure calcined spheres for fissile dosimeters.

The fourth phase is the construction of an EPR1, which should be completed in 2024. A fifth phase of six PWRs resulting from the design of the EPR and currently called “EPR2” is planned for the near future: a first EPR2, construction of which is scheduled to start in 2028 for commissioning scheduled for 2035. For the design of EPR capsules, LDCI recommends reproducing those of the N4, but with the addition of a zirconium dosimeter under thermal filter in order to cover neutron energy between 1 keV and 1 MeV [4].

2 Temporal evolution of ISP dosimeters measurement uncertainties

Figures 1 to 7 show the evolution of the k=1 uncertainty associated with the measurement by the CEA of, respectively, all the uranium, neptunium, nickel, copper, iron dosimeters and those in niobium. For all dosimeters with the exception of the fissile ones, the uncertainty relates to the measurement of the specific activity (Becquerel of the radioisotope of interest per milligram of the dosimeter). The mass of the fissile oxides cannot be directly measurable by the CEA, the uncertainty is therefore that of the total activity (Bq). In these figures, each date that is associated with an uncertainty corresponds by default to the date of the signature of the analysis report. Indeed, the uncertainty value may change between the raw measurement date and the signature of the analysis report. These analysis reports have been systematically under “Cofrac Essais” accreditation since January 1999 for fissile and niobium dosimeters, and since February 1994 for the others. For measurements prior to 2002, the uncertainties were often not provided by the analysis reports but in the interpretation document using the analysis reports (the latter not being signed). In these cases, the date is associated with the start of the count. When the dosimeters of the same type in the same capsule have the same uncertainty value (e.g. the six Al-Co dosimeters of the CPY Chinon B3-V capsule), these dosimeters appear as a single point “X” in the figures.

Each uncertainty value presented in this article aggregates all the uncertainties known at the date of the measurement. Nowadays, these uncertainties contain spectrometry software, photons absorption by the dosimeter itself (and by encapsulation material if fissile), positioning reproducibility on the sensor, reference standard measurements reproducibility, shape (disk, wire, cylinder or tape instead of a point), counting loss due to electronics, gamma-gamma coincidence, and mass for non-fissile dosimeters. Spectrometry software uncertainty includes those related to counting statistics, background noise, counting time, decay during measurement, sensor efficiency, probability of emission of the photon and the correction factor to go back to the end of the irradiation. Some additional uncertainties are specific to the nature of the dosimeter: e.g. for fissile dosimeters, the single escape peak phenomenon due to ^{60}Co when ^{137}Cs is measured, or for niobium, the fluorescence effect or the emission rate of the working standard. Figures 1 to 7 show that between 1981 and mid-1996, the uncertainty associated with a type of dosimeter was an overestimating fixed value (with some exceptions discussed below). These fixed values were determined considering the quadratic sum of the conservative values of the aforementioned uncertainties with the exception of those from: the reproducibility of the measurements of the reference standard, gamma-gamma coincidence or the single escape peak phenomenon due to ^{60}Co when ^{137}Cs is measured. In 1996, this method was considered too conservative and was gradually modified: gradually, each measurement was associated with its dedicated uncertainty (11 successive versions from 1999 to 2023 of the CEA report concerning the expression of activity measurement uncertainties). After 2015, each dosimeter activity measurement uncertainty contains at least three elements, each of which having a ±0.5% minimum uncertainty: the counting loss due to electronics, positioning reproducibility on the sensor, and reference standard measurements reproducibility. A future decrease of one of these items of uncertainties would therefore lead to a generic decrease in future overall uncertainty.

In the rest of this paper, each type of dosimeter will be analysed separately.

Concerning ISP uranium dosimeters, Figure 1 below shows that after 1999 two populations stand out clearly concerning their measurement uncertainty of ^{137}Cs. 1999 corresponds to the year when the CEA ISP measurement platform was moved from Grenoble to Cadarache (before 1999, a fixed value of ±6.7% was used, with only one exception). The reason of two populations among uranium dosimeters is due to the encapsulating material of uranium oxides. The encapsulation by stainless steel (SS) leads to a significant production of ^{60}Co due to the traces of Co inside the steel. The single escape peak due to ^{60}Co leads to polluting the energy peak attributed to ^{137}Cs, up to half of this peak. On the contrary, the encapsulation materials that do not contain Co lead to a lower uncertainty of the activity measured in ^{137}Cs: after 1999, it is always less than ±2.85% with Ni encapsulation and less than ±2.1% with Ti one. The measurement uncertainty of the uranium dosimeter is higher with Ni encapsulation than with Ti due to the ^{60}Ni (n, p) reaction which produces ^{60}Co, even though the single escape peak is lower than the ^{59}Co (n, g) in SS. For several years, a program of re-measurement of the ISP uranium encapsulated in SS, already measured for the first time at least 10 years ago, has been set up in order to benefit from the faster decay of ^{60}Co compared to ^{137}Cs. As this program has not yet been completed, its results are not presented in this article.

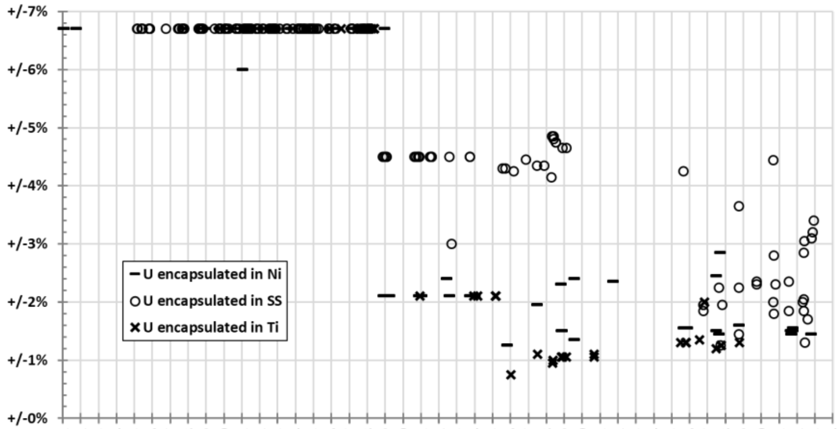

Fig. 1. Evolution over time of the meas. unc. (k=1) of the total ^{137}Cs activity of ^{238}U ISP dosi.

Concerning ISP neptunium dosimeters, Figure 2 below shows that prior to 1999 a fixed value of ±4.0% was used for the measurement uncertainty of ^{137}Cs, except for three dosimeters (these correspond to dosimeters that were badly damaged during extraction, resulting in a high degree of uncertainty about the shape of the dosimeter and the oxide powder density correction factors). The uncertainty values associated with the measurement of these dosimeters have therefore been increased. After 1999, the uncertainty on the total ^{137}Cs activity tends to stabilize around a post-2015 value around ±1.3%, with a few exceptions due to various factors (counting statistics, encapsulation material in nickel leading to a single escape peak phenomenon of ^{60}Co, etc.).

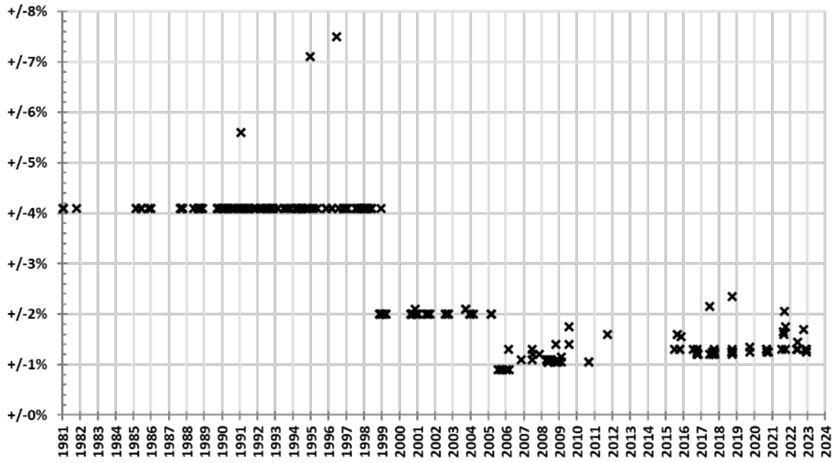

Fig. 2. Evolution over time of the meas. unc. (k=1) of the total ^{137}Cs activity of the ^{237}Np ISP dosi.

Concerning ISP Al-Co bare and shielded dosimeters, Figure 3 below shows that before mid-1996 a fixed value of ±3.0% was used for the measurement uncertainty of ^{60}Co, except for three capsules (i.e. 18 dosimeters) for which it was decided to use a fixed value of ±3.5%. These values included the uncertainty of the cobalt concentration in the Al-Co alloy (main item of uncertainty, up to ±3.0%). After the CEA ISP dosimeter measurement platform was moved in 1999, the uncertainties of ISP Al-Co dosimeters were reassessed by considering only the uncertainties measurable by MADERE: the activity by milligram of Al-Co alloy. The continuous improvement of the dosimeters uncertainties evaluation leads to post-2015 specific activity values of ISP Al-Co dosimeters around ±1.2%.

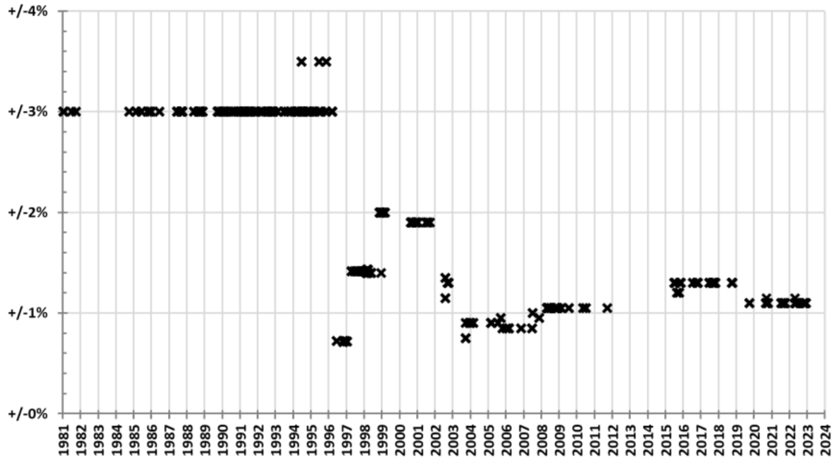

Fig. 3. Evolution over time of the meas. unc. (k=1) of the specific ^{60}Co activity of Al-Co ISP dosi.

Concerning ISP nickel dosimeters, Figure 4 below shows a fixed measurement uncertainty of ^{58}Co before the end of 1995 of ±2.3%, mainly driven by the uncertainty of the sensor efficiency used at that time (i.e. ±2.0% of systematic uncertainty). Drastically higher values for five capsules appear between 1991 and 1998. These specific capsules were sent to

MADERE very late after the end of their irradiation compared to the ^{58}Co half-life: from 15 to 24 periods, whereas the average value without them is 8.6. Since 2015, this value has been even lower: 6.6 in average (ranging from 3 to 10). Measurement uncertainties on the specific activity of ISP nickel dosimeters since 2015 are systematically between ±1.3% and ±1.5%.

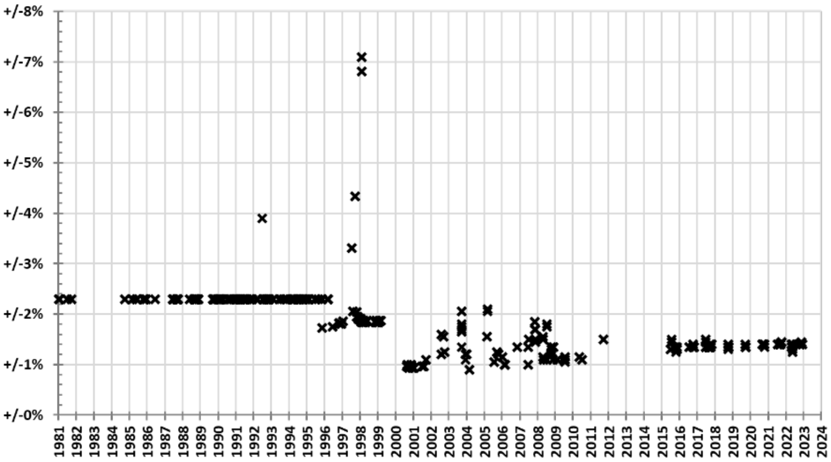

Fig. 4. Evolution over time of the meas. unc. (k=1) of the specific ^{58}Co activity of Ni ISP dosi.

Concerning ISP copper dosimeters, Figure 5 below shows that prior to mid-1996 a fixed measurement uncertainty of ^{60}Co of ±1.8% was used, mainly driven by the uncertainty of the sensor efficiency used at that time (i.e. ±1.7% of systematic uncertainty). After reassessment in 2015, the management of uncertainties leads to measurement uncertainties on the specific activity of ISP copper dosimeters ranging from ±1.2% to ±1.4%.

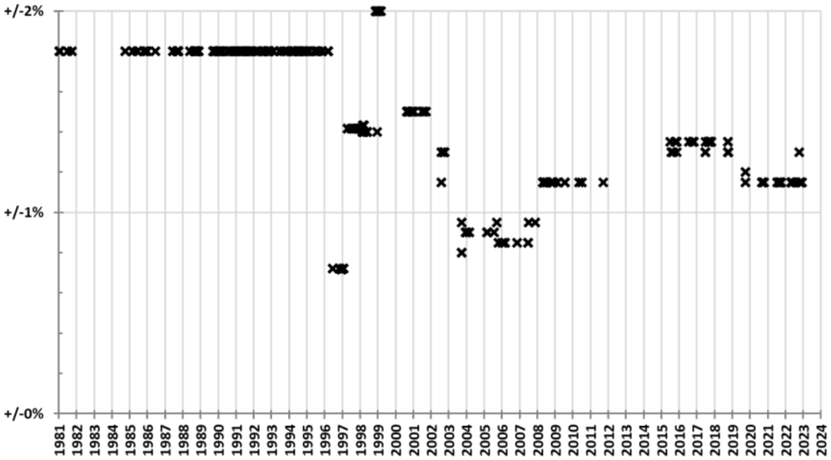

Fig. 5. Evolution over time of the meas. unc. (k=1) of the specific ^{60}Co activity of Cu ISP dosi.

Concerning ISP iron dosimeters, Figure 6 below shows that before end of 1995, a fixed measurement uncertainty of ^{54}Mn of ±2.4% was used, mainly driven by the uncertainty of the sensor efficiency used at that time (i.e. ±1.7% of systematic uncertainty). After reassessment in 2015, the uncertainty management leads to measurement uncertainties on the specific activity of ISP iron dosimeters ranging from ±1.2% to ±1.4%.

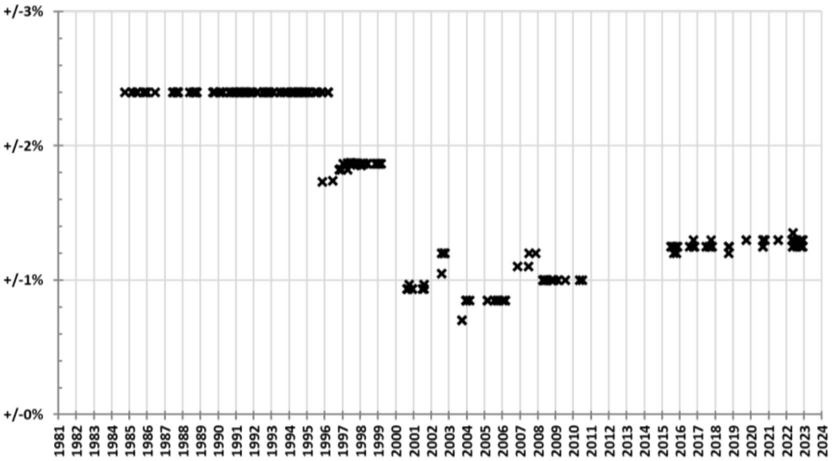

Fig. 6. Evolution over time of the meas. unc. (k=1) of the specific ^{54}Mn activity of Fe ISP dosi.

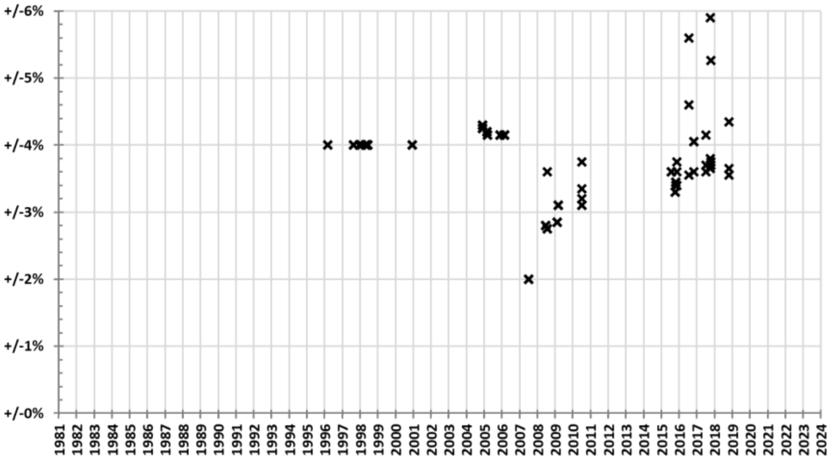

Fig. 7. Evolution over time of the meas. unc. (k=1) of the specific ^{93m}Nb activity of Nb ISP dosi.

Concerning ISP niobium dosimeters, Figure 7 above shows that before 2002 a fixed measurement uncertainty of ^{93m}Nb of ±4.0% was used. Unlike other non-fissile ISP dosimeters, the 2015 reassessment did not lead to a [±1% ; ±2%] range for the measurement uncertainties of ISP niobium dosimeter specific activity. This is due to three causes. First, there is no reference standard for ^{93m}Nb, so a working standard must be used [5]. The working standards available since 2015 have an uncertainty still greater than ±3% (from ±3.1% to ±3.3%). Second, ISP niobium dosimeters must be very thin (20 µm), due to the low emitted photon energies of ^{93m}Nb. This thickness leads to a particularly low mass compared to other ISP dosimeters and therefore to a greater uncertainty associated with mass measurement. Third, this small thickness induces a particular risk of embrittlement during irradiation. Consequently, it regularly happens that ISP niobium dosimeters end up broken after irradiation, resulting in the measurement of dosimeter fragments. For example, each of the ISP niobium dosimeters with an uncertainty greater than ±4.3% systematically corresponds to a broken dosimeter: the largest fragment measured has only from 1/7 to 1/3 of the mass of the dosimeter before irradiation. This phenomenon has led the LDCI to recommend shielding ISP niobium dosimeters to reduce embrittlement during irradiation, e.g. by using cadmium cover. Excluding broken dosimeters, the measurement uncertainty of specific activity of ISP niobium dosimeters ranges since 2015 from ±3.3% to ±3.9%, mainly due to the uncertainty of the working standard.

3 Conclusion and prospects

Four decades of progress in measurement techniques, nuclear data and quality management systems have made it possible to reduce the uncertainties associated with activity measurements of ISP dosimeters. Typical 1σ measurement uncertainties since 2015 of ISP uranium dosimeters activities are [±1.3% ; ±2.9%] with nickel encapsulation, [±0.8% ; ±2.1%] with titanium encapsulation and [±1.2% ; ±4.9%] with stainless steel encapsulation, ±1.3% for neptunium dosimeters, ±1.2% for Al-Co, [±1.2% ; ±1.4%] for nickel, copper and iron, [±3.3% ; ±3.9%] for niobium if unbroken after irradiation. Thus, apart from niobium, the standard uncertainty has been divided by an approximate factor of 1.4 for copper to 3 for neptunium since 1981. These reductions enable consolidation of the neutron fluence values determined by LDCI and provided to EDF to assess PWRs vessels irradiation embrittlement, which is crucial for obtaining the right by the Nuclear Safety Authority to extend their operating life.

Several LDCI R&D programs are underway to continue to progress on the best assessment of the measurement uncertainty of activity, such as absolute measurement of niobium dosimeters to avoid the use of uncertain working standards and Monte Carlo modelling of detectors used for gamma and X-ray spectrometry [6].

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