2014 CALIBAN AND PROSPERO EXPERIMENTS FOR THE CRITICALITY ACCIDENT DOSIMETRY INTERCOMPARISON

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ABSTRACT

Nuclear criticality accident dosimetry is very specific, due to the mixed fields and to energy and dose ranges which could be out of the range of classical individual dosimetry. The maximal neutron and gamma dose must be determined as well as other information such as the orientation of the victim in the radiation field. It is exceptionally challenging to establish doses in the event of partial body exposures. Therefore, frequent exercises are organized to maintain competences and to train staff to perform such dosimetry.

Experiments using the CALIBAN and PROSPERO reactors were performed in June and September 2014 in order to test various criticality accident dosimeters (CAD). These two metallic reactors are located at the CEA research center at Valduc (Côte-d'Or, France). They are composed of a metallic alloy of uranium with a high enrichment in the isotope ^{235}U. These reactors are used for the production of mixed radiation fields primarily comprised of fission neutrons and gammas.

CADs from three different laboratories (AWE, UK ; IRSN, FR ; LLNL, USA) were irradiated on water phantoms with three different orientations (0° and 45°, front and rear faces) and free in air at both CALIBAN and PROSPERO reactors, in pulse and steady state modes. This paper will present the comparison of some results. These experiments will be useful preparation for other exercises planned in 2016 at the GODIVA-IV and FLATTOP reactors located at the Nevada National Security Site, USA.

KEYWORDS
Criticality accident, dosimetry, intercomparison

1. INTRODUCTION

Since 1945 about sixty criticality accidents were reported [1] [2]. Most of these accidents occurred in the fifties and sixties during the age of development of nuclear weapons and nuclear reactors with lower standards of safety and more intense research activities than today. Over the last 20 years, only three accidents have occurred. This typology of accident is less frequent than accidents in radiotherapy or in other radiation related work; however the consequences can be much more severe. The last two accidents led to the death of 3 of the 4 exposed personnel. Over the years, approximately 21 persons *in toto* have died from criticality accidents. The high level of safety in nuclear facilities today reduces the criticality risk, but the possibility of an accident cannot be excluded as it was dramatically demonstrated by the latest accident in Japan in 1999 [3].

Conventional dosimeters used for routine monitoring of workers cannot adequately estimate dose in the event of a criticality accident. The dose may exceed the dose range of conventional dosimeters especially for the neutron components, the dose rate can be very high, and it is necessary to separate photon and neutron doses. It is also very important to determine the orientation of the exposed personnel during the event in order to assess the maximum dose and dose distribution in the body or dose to the most critical organs. Moreover, when a criticality accident occurs, it is necessary to rapidly identify the highly exposed people so that they can be directed to medical treatment. Preliminary dose assessments for each exposure individual must be performed rapidly within 24 hours following the accident [4]. In addition the dose quantities which have to be reported for criticality accidents are not the same than for routine dosimetry. Sievert (Sv) does not have any signification in this context, and quantities are reported in terms of Gy [4] [5]. All these constraints led to the development of specific dosimeters and approaches, as well as a different evaluation regime than for routine individual monitoring. Often times whole dosimetry systems with different components have been developed specifically for criticality dosimetry. As an example, the gold standard system recommended by IAEA [5] and as developed in the 1960s was ideally composed of: 1. a locket attached to the routine dosimeter; 2. a belt with several detectors for the orientation and; 3. an area dosimeter that incorporates a neutron spectrometer based on activation. In neutron dosimetry, the knowledge of the neutron spectra is an advantage in providing accurate dose estimation, due to the variety of neutron interactions that depend on neutron energy. However it becomes problematic in determining the neutron spectrum after an accident. So, the criticality dosimeter design must be able to properly respond regardless of spectral changes or provide an estimate of the neutron spectrum so that accurate doses can be determined. One important additional consideration is that these dosimeters are only analyzed in case of accident. Therefore, it was also important to minimize the maintenance needs of the dosimetric components.

2. SPECIFICITY IN THE MANAGEMENT OF CRITICALITY DOSIMETRY SYSTEMS

Many criticality dosimetry systems use neutron activation techniques to assess neutron doses. Neutron dose quantities can be determined by evaluating the activities of activated metallic foils. Some of the foils have threshold energies for neutron activation while others may require shielding to limit the response of a foil to a specific range of neutron energies. This approach allows estimating separately dose components from different neutron spectra. Moreover this technique is not sensitive to dose rate and it requires minimal maintenance. Typical metallic foils currently used in these systems include gold, indium, sulfur and copper. However, the short half-lives of some of the induced radionuclides require that the measurements of the activation be performed with minimal delay. This constraint requires, in terms of organization, that the measurements be performed as close to the accident site as reasonably possible and not in an individual monitoring laboratory at large distances from the event. It means that on each nuclear site, some personal have to train to analyze the dosimeters and to evaluate dose. As the dosimeters are not routinely analyzed, regular exercises should be organized to maintain competences and train new personnel as well as to check for the proper functioning of the instrumentation. The main difficulty for the institutions where criticality accidents could occur is to maintain competencies and an operational team for an event which has a very low probability of occurrence, but where the consequences are severe and for which it is important to react immediately. For these reasons it is important not only to organize frequent exercises to train staff and keep instrumentation functioning, but also to compare the different dosimeters in order to improve the quality of the dose assessments.

This is not only true for the analysis of criticality accident dosimeters, but also for staff in charge of quickly sorting potentially exposed personnel and staff in charge of analysis of activated biological samples in biomedical laboratories.

It is also necessary to be able to assess the doses even if no dosimeters are worn. Therefore "retrospective" dosimetry techniques have also been developed based on analysis of biological samples and other materials that may be available. Irradiation of a person results in activation of biochemicals containing sodium or sulfur. Blood sampling or whole body counting for sodium or sulfur activation in hair and nails are useful bioassay techniques for determining the dose to personnel from criticality accident exposure. Cytogenetic assay, if proper calibration curves for mixed radiation fields have been established by the laboratory, can also be used as a mean to assess the dose. More recently, techniques based on quantification by Electron Paramagnetic Resonance (EPR) spectroscopy of free radicals induced by photon irradiation can be used to estimate dose in nails or in mini-biopsy of tooth enamel [6] [7]. All these retrospective techniques were not tested during the intercomparison of September 2014 at Valduc and are therefore outside of the scope of this paper. It will not be discussed or described further.

To conclude, since criticality accidents are nowadays very rare, the preparedness for these types of accidents is all the more challenging. Staff must be trained to react, in a very short delay, to perform victim triage, potential decontamination, dosimetry measurements, biological assay, retrospective dosimetry evaluations, and more depending on the nature of the event.

3. ACCESS TO CRITICALITY FACILITIES AND INTERNATIONAL COLLABORATION

3.1. The most recent intercomparisons and need for new ones

Irradiations were usually performed around experimental reactors in order to irradiate dosimeters with sufficient doses (especially neutron dose) and with energy distributions close to those encountered at a real accident to test dosimeters and train personnel. Maintaining experimental reactor facilities can be very hard to support for one single institute. It is worth noting that the list of available facilities in the world has become shorter in recent years. These difficulties had been acknowledged by the IAEA who organized, in the nineteen seventies, a series of four criticality accident exercises using four different facilities [8] [9] [10] [11]. The CALIBAN and PROSPERO reactors (Valduc, France) used for this intercomparison are now definitely closed since December 2014. It follows the closure of SILENE reactor (Valduc, France) in 2010. Organizing criticality dosimetry training and intercomparison exercises in Europe in the future has now become impossible. The possibility to organize future international intercomparisons is also somewhat uncertain. It is worth noting that the three last international intercomparisons were organized at CEA facilities: on SILENE in 1993 [12] and 2002 [13] and on CALIBAN in 2010 after the restart of the reactor in 2007 [14] [15]. The last published comparison of results was from the 2002 intercomparison. The 2002 campaign was organized with the help of the NEA/OECD and was partly sponsored by the European Communities. The intercomparison involved a total of 60 laboratories from 29 countries and provided participants with a practical training opportunity to test their dosimetry systems under realistic conditions for three different quality radiation fields. The overall analysis concludes with fairly good agreement between the measurement results and reference values for the neutron and gamma rays [13]. The exercise also demonstrated the absolute necessity to perform periodic tests of criticality accident dosimetry. Indeed, intercomparisons gave the participants an opportunity to improve their techniques of measurements and to evaluate the performance of their dosimetry systems through controlled irradiations in well-characterized fields of radiations. It is also an indispensable way to train staff to perform criticality dosimetry in the event of an accident.

The possibility to maintain operational competences on every nuclear site will become a problem in near future. It is especially the case in France and that is why IRSN has proposed a new dosimeter to its customers which requires minimal training and effort for analysis. However, even with improved dosimetry systems, there is still a need for national and international collaboration and cooperation in this specific field of dosimetry to maintain and to share the few facilities still available and to have scientific cooperation for future developments.

3.2. The AWE-IRSN-LLNL collaboration

The MIDAS project was initiated in collaboration with the US DOE/NNSA, IRSN and CEA (France) and consisted of the refurbishment of the 010 building in the CEA/Valduc as well as the creation of a new SILENE reactor and “Apparatus B” subcritical assembly. The MIDAS project was to develop the next available intercomparison facility, but was discontinued in 2013. IRSN has launched the PRINCESS project (PRoject for IRSN Neutron physics and Criticality Experimental data Supporting Safety), which aims at the acquisition of experimental data in the field of criticality safety and reactor physics. This data might be obtained by several ways, i.e. data acquisition from finished programs, collaboration on ongoing programs or collaboration on future programs with different ways of participation (funding, needs analysis, experimental design, results analysis…) and with international partners from USA, Japan, Russia, Europe, OECD/NEA, IAEA, etc.

The intercomparison presented in this paper is one of the first steps to mutualize and coordinate efforts between UK (AWE), USA (LLNL) and France (IRSN). IRSN invited LLNL and AWE to test criticality accident dosimeters (CADs) and other devices during the last IRSN experiments with CALIBAN and PROSPERO reactors (which are similar to GODIVA-IV and FLATTOP reactors). AWE and IRSN were also invited by the US DOE/LLNL, in the framework of the US Nuclear Criticality Safety Program (NCSP) [16], to participate to various experiments with GODIVA-IV and FLATTOP reactors. The first action was the participation of AWE and IRSN to the characterization of GODIVA-IV radiation field in 2014 (IER 147).

The experiments presented in this paper took place in September 2014 at the CEA research center at Valduc (Côte-d'Or, France) with CALIBAN and PROSPERO reactors in order to test various CADs in different conditions. The three laboratories, AWE, IRSN and LLNL, were involved and exposed their CADs on water phantoms with different orientations (0° and 45°, front and rear faces) or station free in air. Preliminary measurements and field characterization (neutron spectrometry) were done by AWE and IRSN in June 2014 at CALIBAN and PROSPERO reactors.

4. MATERIALS AND METHODS

4.1. Description of the CALIBAN and PROSPERO reactors

Few facilities in the world can produce intense radiation fields simulating accident conditions. In France, three reactors, situated at Valduc (Côte d’Or) and operated by CEA, could do so: the SILENE reactor, the CALIBAN reactor and the PROSPERO reactor. The SILENE reactor was closed at the end of 2010 and the CALIBAN and PROSPERO reactors were decommissioned at the end of 2014 shortly after the AWE, IRSN, and LLNL measurements.

During the intercomparison of September 2014, the irradiations were performed at both CALIBAN and PROSPERO reactors. They are usually dedicated to the study of the hardening of components and electronic systems but they are also used for the production of neutrons and gammas of fission. They are composed of a metallic alloy containing uranium with a high enrichment in isotope ^{235}U. Both reactors are located in large concrete cells: 10 x 8 x 6.5 m^3 for PROSPERO and 10 x 8 x 5 m^3 for CALIBAN.

4.1.1. The CALIBAN reactor

The CALIBAN reactor core is a cylinder shape with a vertical axis. This experimental reactor consists of a solid core made of 10 fuel discs and 4 rods of 93.5 % enriched uranium metal alloyed with 10 wt% of molybdenum, with a combined weight of 113 kg. Additional information is given in reference [17].
The reactor was used in pulsed mode; a typical CALIBAN pulse reaches a 20 GW power with a full width at half maximum of 50 µs. During the experiment of September, about $1{,}98.10^{16}$ fissions were produced.

4.1.2. The PROSPERO reactor

The PROSPERO reactor core is a cylinder shape with a horizontal axis. It is surrounded by a depleted uranium reflector. This reactor usually works in a steady-state mode and can reach a 3 kW maximal power. During the experiment of September, about $5{,}3.10^{15}$ fissions were produced.

4.2. Dose quantities

The dose quantities and the information that are reported in this manuscript are specific to criticality accident dosimetry. The maximum absorbed doses (neutron and gamma) in the body were evaluated. For neutrons, the dose is due to heavy charged particles, to protons from $^{14}N(n,p)^{14}C$ reaction and to gamma rays from $^{1}H(n,\gamma)^{2}D$ reaction. These components of the dose have been calculated by Auxier, et.al. in the « Element 57 » of a cylindrical phantom model [18]. The dose due to protons from $^{14}N(n,p)^{14}C$ reaction in tissue is usually estimated by measuring the neutron kerma in tissue. When dosimeters are worn, it may be necessary to correct the reading to estimate this quantity as it was measured in real kerma conditions (free in air). In addition, the assessment of the gamma dose may be complicated by the response of gamma dosimeters to neutrons which has to be known to perform a correction on the so called “direct gamma dose” which is the maximal dose absorbed in the tissue.

According to IAEA Technical Report #211 [5], for dosimeters exposed on phantoms the quantities involved are:

- D_n, neutron recoil + proton dose for Element 57,
- $D_{n\gamma,}$ neutron H(n-γ) dose for Element 57,
- $D_{t\gamma,}$ total gamma ray dose (incident dose + (n-γ) dose).

For dosimeters exposed in free-air, the principal quantities involved are:

- $K_{n,}$ neutron kerma,
- $D_{\gamma,}$ incident gamma ray dose,
- and the neutron fluences above given energy limits.

5. CRITICALITY ACCIDENT DOSIMETERS

5.1. AWE system

The AWE CAD is the Harwell Mk III neutron dosimeter, developed in the 1960s by the UK Atomic Energy Agency and was placed in close proximity to CALIBAN and PROSPERO reactors. The elements of the locket consist of sulfur, indium, gold, cadmium and plastic. AWE was testing a new method for the interpretation of this dosimeter, which is described elsewhere in these proceedings [19]. Photon measurements were performed with a Harshaw Type 8814 thermo-luminescent dosimeter (TLD), which was mounted close to the locket.

5.2. IRSN system

IRSN deployed several dosimetric systems:

- SNAC2, an area dosimeter which is a neutron spectrometer with activation foils of copper, gold, indium, magnesium and nickel,
- New IRSN CAD, including alanine pellets and RPL dosimeter,
- Silicon diodes,
- Sulfur pellets.

The photon component was assessed with RPL glasses. The passive silicon diodes, the activation detectors and alanine pellets measured by electron paramagnetic resonance (EPR) spectroscopy gave the neutron component.

5.3. LLNL system

The LLNL Personnel Nuclear Accident Dosimeter (PNAD) consists of a Panasonic 810 TLD, gold, indium and copper foils, a sulfur pellet, with cadmium and borated plastic shields, and plastic caps inside a plastic case. Fixed Nuclear Accident Dosimeters (FNADs) contain the same foils and shields as the PNAD, only larger. Panasonic 810 TLDs were placed on top or in proximity to the FNADs. LLNL also used Personnel Ion Chambers (PICs) for the photon doses.

6. EXPERIMENTS PERFORMED

6.1. Measurement locations

The CADs were placed on phantoms or on an aluminum backed station free in air. The phantoms were filled with water. The phantoms' stands were located at 80 cm above the ground. The orientation of the majority of the phantoms was 0° compared to the reactor (anterior-posterior orientation). But in order to study the influence of the orientation of the phantom, some of them were rotated 45° degrees relative to the direction of the assembly. The figure 1 shows the experimental set-up at CALIBAN reactor.
For the dosimetric measurements, there were one station free in air and two phantoms situated at 3 m from the CALIBAN reactor. For the PROSERO exposure, two phantoms and one station free in air were used, all situated at 3.5 m from the core. The influence of phantom orientation and of the different radiation fields was studied. Moreover, the dosimeters were mounted on the front and rear faces of the phantoms.

6.2. Proceedings

Due to difficulties accessing the CEA center, people from AWE and LLNL could not be present at Valduc for the irradiations. IRSN members performed the experiments placing the dosimetric systems in the different configurations. AWE and LLNL participants brought their own equipment for the measurements and assessment of their dosimeters at the IRSN site situated at Fontenay-aux-Roses (Paris). This included gamma ray spectrometers and beta counting equipment. All the IRSN equipment to analyze the dosimeters are at the IRSN laboratory site, except the RPL readers which are situated at the Radiation Monitoring Department of IRSN at Croissy-sur-Seine (Paris). Once the irradiations were completed with the reactors, the dosimeters were transported to the laboratory at Fontenay-aux-Roses where the first measurements, especially measurements of activated foils, could be performed*. On their return home,

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* Between the end of the irradiation and the start of measurements, about 7 hours were necessary to leave the CEA center and transport the dosimeters.

participants have completed assessment of dosimeters (by reading the thermoluminescent or RPL detectors for instance) and the obtained results were sent to the organizers.

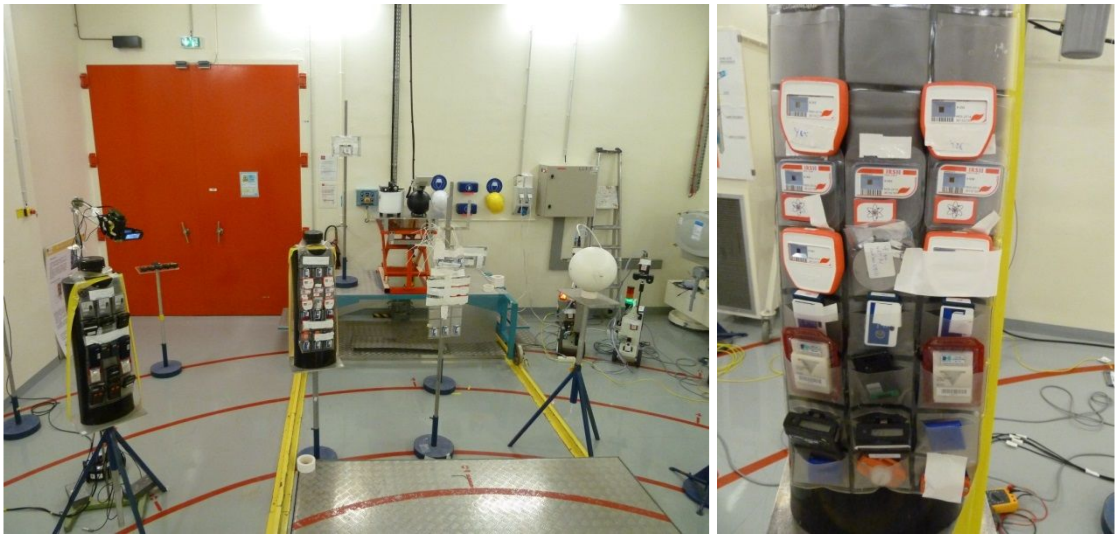

Figure 1 On the left, CALIBAN reactor – September 3rd experimental configuration; on the right, phantom with the different CADs.

7. MEASUREMENT AND EVALUATION OF DOSIMETERS – RESULTS

The dose estimates supplied by participants were derived from the measurements of their exposed dosimeters. The photon dose corresponds to absorbed dose and the neutron dose is given in neutron kerma in tissue. The activation foils and the silicon diodes were measured within 12 hours after the irradiation, whereas other types of dosimeters (mainly TLD, RPL and alanine) were read out in each laboratory within two weeks following irradiation. Obviously, this is not representative of actual time response of each laboratory. As the readers used are those used for routine individual monitoring that was impossible and not necessary to move all these cumbersome and expensive equipment. In real situation as the dosimeters are transported directly to each laboratory, the reading can be performed in short delay.

7.1. Neutron kerma in tissue

The Tables I and II give the neutron kerma in tissue from the different laboratories for the station free in air and phantom at 0° and front side at each reactor.

Table I. Neutron kerma in tissue in Gy for the CALIBAN irradiation, at 3 m and 0°.

phantom or stand		**Reference value (free in air)**	**AWE**	**IRSN (alanine)**	**IRSN (diode)**	**IRSN (SNAC)**	**LLNL**
stand	free in air	1.17	-	1.39	1.29	1.07	1.11
phantom	front		1.19	1.34	1.30		1.20
Relative uncertainties		~10 %	~10 %	~15 %	~10 %	~20 %	~10 %

Table II. Neutron kerma in tissue in Gy for the PROSPERO irradiation at 3.5 m and 0°.

phantom or stand		Reference value (free in air)	AWE	IRSN (sulfur)	IRSN (diode)	IRSN (SNAC)	LLNL
stand	free in air	0.12	-	0.12	<DL	0.10	0.13
phantom	front		0.13	0.11	<DL		0.13
Relative uncertainties		~10 %	~10 %	~18 %		~20 %	~10 %

There was good agreement between the three laboratories for both CALIBAN and PROSPERO measurements considering the uncertainties. Moreover, all the neutron doses are within 10 % compared to the reference value, except for the measurements performed with alanine pellets for the CALIBAN irradiation for which an overestimation of 20 % is observed. The reference values were determined in the cell emptied of water phantoms and any scattering materials. The influence of these scattering materials has to be evaluated. Simulations to estimate the actual influences of the phantoms and free in air stand are progress and may be presented at the conference. The measurement with the neutron IRSN spectrometer SNAC2 underestimates the reference value by 15 % for the PROSPERO irradiation. This can easily be understood by the chosen method of unfolding. Indeed, the development of this spectrometer and the associated analysis were based on test performance around the SILENE liquid reactor which presented an important contribution of the thermal neutrons. For neutron spectra presenting a lower thermal fluence such as the ones encountered at CALIBAN and PROSPERO facilities, the unfolding code tends to underestimate the epithermal portion of the spectra. This phenomenon was previously shown in measurements at the CALIBAN reactor [17]. To solve this problem, a new version of the unfolding code is currently being developed at IRSN. For LLNL and IRSN dosimetry systems (no measurement for AWE), the differences between the measurements performed on phantom and station free in air are within 10 %. This demonstrates that the system designs allow for measurements on the body while obtaining a correct estimate of the neutron kerma. The neutron doses were too low for the PROSPERO irradiations to be measured with silicon diodes. The doses were below the detection limit.

7.2. Photon doses

The Tables III and IV give the absorbed doses for photons from the different laboratories for the station free in air and phantom at 0° and front side at each reactor.

Table III. Photon doses in Gy for the CALIBAN irradiation, at 3 m and 0°.

phantom or stand		Reference value (free in air)	AWE (TLD)	IRSN (RPL)	LLNL (TLD)	LLNL (PIC)
stand	free in air	0.22	-	0.22	0.44	0.53
phantom	front		0.40	0.31	0.66	0.56
Relative uncertainties		~10 %	~10 %	~10 %		

Table IV. Photons doses in Gy for the PROSPERO irradiation at 3.5 m and 0°.

phantom or stand		Reference value (free in air)	AWE (TLD)	IRSN (RPL)	LLNL (TLD)	LLNL (PIC)
stand	free in air	0.03	0.04	0.04	0.05	0.12
phantom	front		0.07	0.06	0.09	0.15
Relative uncertainties		~10 %	~[12-14] %	~10 %		

Although the techniques used is completely different (TLD or RPL), there is a good agreement between AWE and IRSN for the absorbed dose for photons for all the configurations. For the CALIBAN irradiation, we noticed some differences with LLNL photon doses measured with TLD and Personnel Ion Chamber (PIC) readings by as much as a factor 2. This result has yet to be understood. For the PROSPERO irradiation, the LLNL measurements with the TLD are similar to those from AWE and IRSN. Only the PIC readings provide higher values of doses. This result has yet to be investigated.

8. THE NEXT EXPERIMENTS

A new intercomparison is planned in 2016 at the GODIVA-IV reactor at the Nevada National Security Site (NNSS), USA within the American Integral Experiment Request 148. It will be the first intercomparison after the transfer of GODIVA-IV from Los Alamos National Laboratory to the NNSS. The characterization of the reactor was performed in 2014, with spectrometric and dosimetric measurements [20]. Compared to the intercomparison performed at Valduc in September 2014, many more laboratories will be involved. In addition to LLNL, AWE, and IRSN, eight other US laboratories will participate. The first irradiations will be performed to allow laboratories to test their dosimetry systems, train, and compare to known values. The final irradiation of the intercomparison will not only provide a technical comparison of the different systems to a reference dose only known to the operators of the exercise, but will also emphasize the operational aspect: the laboratories will have to give a first estimation of the dose within the first 24 h after the irradiation. Another comparison on the FLATTOP reactor is in the preplanning stages and could also have intercomparison exercises after its characterization occurring in 2016.

9. CONCLUSIONS AND PERSPECTIVES

This exercise was a first step of the collaboration between AWE, IRSN and LLNL and establishes a path forward for future intercomparisons. Above all, this exercise enabled training of personnel who have little opportunity and operational experience at performing criticality dosimetry measurements. It was also an opportunity to test several unique systems in well-known radiation fields before the decommissioning of the facilities of Valduc. The results showed good agreement among the principal configurations. Some investigations and additional tests or simulations have to be done to understand some of the observed disparities. Such exercise has to be repeated in order to test CADs performance in other configurations. Finally these experiments were very useful for the preparation of the new intercomparison planned in 2016 at the GODIVA-IV reactor.

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