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Neutron personal dosimetry using polyallyl diglycol carbonate (PADC): Current status, best practices and proposed research

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ABSTRACT

The objective of this paper is to provide an overview of the current status in neutron personal dosimetry based on poly allyl diglycol carbonate (PADC), also commonly known by the commercial name CR-39, to summarize the best practices in the field, and to point future research directions. An overview of the fundamentals of the technique is given, including a discussion on the PADC material, main parameters and characteristics, practical considerations, dosimetry approaches, and relevant standards. This work also summarizes the best practices adopted by individual monitoring services (IMSs) and discusses the research needed to improve the performance of this type of neutron dosimetry technique, as well as the challenges that make progress difficult. This work is based on the knowledge and experience of several laboratories and investigators and is part of the activities of the European Radiation Dosimetry Group (EURADOS) Working Group 2 –Harmonization of Individual Monitoring in Europe (WG2).

1. Introduction

Individual monitoring of neutron exposures is challenging due to the complexity of neutron detection: the extremely wide energy range of the neutrons to be considered (nearly ten orders of magnitude), the high variability of spectra that can be encountered at workplaces, the fact that neutron detection can only be done by means of secondary charged particles, and the systematic presence of an associated photon field, which requires appropriate discrimination in case of neutron dosimeters that are also sensitive to photons [43].

Measurements with reference spectrometry in the frame of the EVIDOS project revealed significant differences in the energy distributions of workplace neutron fields [52]. All distributions, however, exhibit a similar structure: a thermal peak, a rather flat intermediate-energy region (in lethargy representation), and a high-energy peak with a maximum between 100 keV and 10 MeV. Neutrons with even higher energy can be found, for example, at high energy accelerators [11,84]. The importance of the different neutron energies for dosimetry depends on the fluence-to-dose equivalent conversion coefficients $h_p(10)$, which vary by a factor of about 50 across the energy range of practical interest for most workplaces [42]. Because of this strong energy dependence, the contributions of high-energy neutrons to dose equivalent is dominant in all spectra [52].

It is therefore important to know the response function of the dosimeter as a function of neutron energy, or - better - to have an energy response as flat as possible in the energy range of interest, for the quantity of interest. This is all the more justified as, in practice, the reference fields used for the initial characterization and routine calibration of dosimeters do not correspond to the spectra encountered in the workplace. The neutron sources which are easily accessible have an energy distribution that is shifted towards high energies. Although the energy response can be characterized using monoenergetic neutron fields, these irradiations are expensive and a set of irradiations at different energies is required to cover the typical neutron energy spectra

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met in practice. Similarly, the dependence of the response on the angle of neutron incidence is a critical aspect which must be properly investigated. Such issues are highlighted by the revision of the ISO 21909 standard, which requires many tests with different neutron fields to carefully characterize the energy and angular dependence of the response [48]. The difficulty of neutron dosimetry is further enhanced by the fact that the workplace dose levels to be measured are near the dosimeter's detection limit.

Poly allyl diglycol carbonate (PADC), also known by the brand name CR-39, is a plastic polymer that has become a standard Solid-State Nuclear Track Detector (SSNTD) for the detection and dosimetry of ionizing particles [18] and is used routinely in radon monitoring and neutron dosimetry. PADC-based neutron dosimeters are among the most widespread devices used for neutron personal dosimetry, as they are simple and inexpensive passive dosimeters which are suitable for a wide range of practical workplaces.

The signal recorded by PADC consists of tracks resulting from damage to the material polymer chains, which are enlarged by a treatment known as *etching* and can be visualized using an optical microscope and counted [31] (see Fig. 1). In the case of neutron detectors, damage trails are produced by neutron-induced secondary charged particles, generated by the interactions of neutrons with the detector itself or the materials surrounding it, with sufficiently high energy deposition per unit path length (linear energy transfer, LET). These are mainly recoil protons from elastic scattering of neutrons with hydrogen. PADC detects radiation with LET values greater than approximately 10 keV/μm, being insensitive to beta and gamma radiation [51]. The damage trails corresponding to the breaking of chemical bonds in the polymeric chains of PADC due to ionization events are known as *latent tracks*, which become proper tracks visible under the optical microscope after etching.

Two types of etching procedures are used: chemical etching (CE) and electrochemical etching (ECE). In CE, the detectors are immersed in the etching solution (etchant), typically a NaOH or KOH aqueous solution, which erodes the damaged material at a rate greater than the undamaged material, so that tracks with a conical shape are produced [61]. In ECE, an electric field is used to enhance the erosion leading to tree-shaped tracks [3,67,81].

Nevertheless, there are open issues in PADC neutron dosimetry, mainly linked to the material quality of the detector itself, which compromise its performance and application, including:

a) high and variable intrinsic background (i.e., signal registered on unexposed detectors) across PADC sheets/batches;
b) variable sensitivity across PADC sheets/batches; and
c) worsening of the PADC sensitivity in time, linked to the *ageing* and *fading* phenomena (see Section 2.2.8), varying also from batch to batch.

These problems were highlighted in a survey of Individual Monitoring Services (IMSs) in Europe carried out in 2012 by the European Radiation Dosimetry Group (EURADOS) Working Group 2 – Harmonization of Individual Monitoring in Europe (WG2) [34]. The survey shows that one common source of error for PADC-based neutron dosimeters is the variability of material quality, in addition to unstable etching conditions. The variable material quality of the PADC detectors impacts their detection limit and performances at very low dose levels.

Because of the issues above, guaranteeing the quality of the PADC measurements in a neutron dosimetry service remains challenging and time intensive. Different laboratories have gained decades of experience with the technique and have developed their own quality assurance procedures, but this knowledge remains scattered and often not documented in the literature. Progress to improve the technique, either for better dose assessment or to simplify the quality assurance programs, has been slow. Therefore, there is a need for a more coordinated effort to assess the current status of the technique and decide, based on the experience of different laboratories, what the most sensible and practical steps to better understand and improve the technique would be.

With that in mind, this work aims at providing an overview of the current status of neutron personal dosimetry based on PADC, summarizing the best practices based on the experience of the various participating European laboratories and using this combined knowledge to try to identify the most relevant areas of investigation. This paper is the first step of a new task of the EURADOS WG2 whose objective is to improve the quality, and thus the performance, of personal PADC neutron dosimeters and to disseminate the best practices related to their use.

2. Overview of the technique

2.1. The PADC detector

Poly allyl diglycol carbonate (PADC), with formula $(C_{12}H_{18}O_7)_n$, is a clear thermoset resin with density of 1.31 g cm^{-3} and essentially transparent to visible light (Fig. 2) [19]. The polymer was patented in the 1940s with the brand name CR-39® that stands for "Columbia Resin #39", since it was the 39th polymer (out of about 200) developed by the Columbia-Southern Chemical Corporation (now Pittsburgh Plate Glass Industries, PPG) in the frame of the *Columbia Resins Project* [12]. The material was identified as a solid-state nuclear track detector (SSNTD) by Ref. [18] and it soon became one of the most widespread track detectors. The name CR-39 has become a synonym of PADC, even when the polymer is produced differently from the original CR-39® patented by PPG; for this reason, the term PADC is more general and should be preferred.

PADC is made by the polymerization of the monomer allyl diglycol carbonate (ADC) (Fig. 2), in the presence of an initiator. This initiator is typically diisopropyl peroxydicarbonate (IPP) [78]. Alternatives are cyclohexyl peroxycarbonates (CHPC) and benzoyl peroxide (BPO) [59]. The presence of the double bonds in the allyl groups ($-CH=CH_2$) of

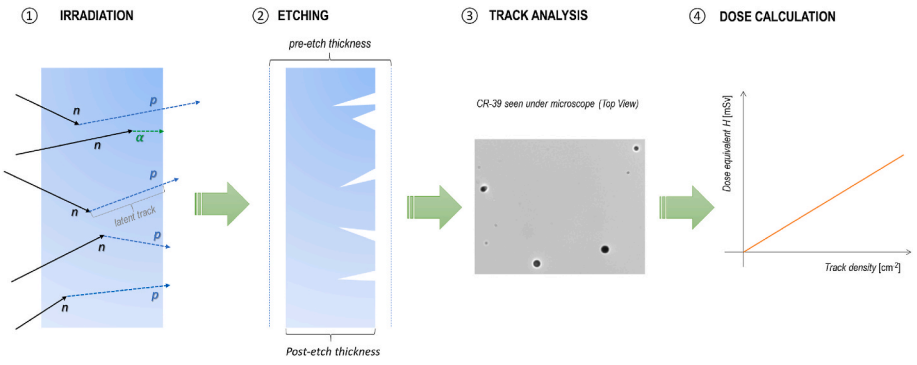
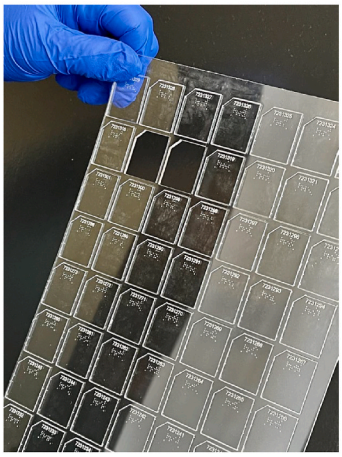


Fig. 1. Main steps of neutron dosimetry using PADC for a dosimeter consisting of a bare PADC detector: ① irradiation: neutrons interact with PADC generating charged secondaries, which ionize the material breaking its polymer chains, producing latent tracks (neutrons scattered after the interactions are not shown); ② etching: the irradiated PADC undergoes etching (chemical etching, in this example); ③ track analysis: the etched PADC is observed under the microscope (20 × objective, for the frame reported), where tracks appear as black ellipses, and tracks are analysed (for example, they are counted and divided by the scanned area to obtain the track density); and ④ dose calculation: the dose is estimated on the basis of the track analysis (by converting the track density in dose equivalent through a conversion factor, in this example).

(a) (b)



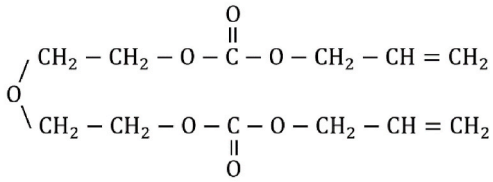


Fig. 2. (a) Example of PADC sheet and (b) chemical structure of PADC monomer (ADC).

the monomer allows the polymer to form cross-links, resulting in a thermosetting resin. The conditions used for this polymerization are termed *cure cycle*.

Various PADC cure cycles have been developed, although most details remain unpublished for commercial reasons. Noteworthy cure cycles are the original controlled PADC cure cycle described by Ref. [23]; its modification by Ref. [1]; leading to a more consistent PADC, and the cure cycles by Ref. [2] to produce an optimized PADC with the highest density of cross-links. The polymerization schedule of PADC monomers using IPP is generally 20 h long, with a maximum temperature for the mould of 95 °C, elevated temperatures being handled with water bath or forced air oven [24].

The PADC material as prepared above is optically transparent, highly isotropic and homogeneous, very sensitive to ionizing radiation, shows no repair of cross-linking after radiation damage breaks the chemical bonds, and has a non-solvent chemical etchant, i.e. the etchant degrades the polymer instead of dissolving the material into solution [18,19]. Moreover, when used as a SSNTD, plasticizers can be added to PADC to avoid the "fogging" caused by the chemical etching, and antioxidants can be added to lower the minimum LET needed for track formation [80].

PADCs produced by different suppliers may differ with regards to Ref. [80]:

1. monomer supply (dimer or trimer);
2. temperature and duration of the cure cycle;
3. curing orientation (horizontal *versus* vertical);
4. initiator (type and concentration); and
5. additives (antioxidants, plasticizers, etc.).

These factors affect the performance of PADC used as nuclear track detector (e.g. sensitivity to radiation, signal to noise ratio, intrinsic background, etc.), as highlighted by several studies [2,32,41,68,70,83].

2.2. Main parameters and characteristics

The physics of track formation in SSNTDs has been described in several publications [31,40,60,61,63,66,69,71,87]. The main parameters are here summarized.

2.2.1. Bulk etch rate V_b and removed layer h

The bulk etch rate V_b (μm h^{-1}) is the speed at which the etching solution erodes the undamaged material, i.e. the bulk (Fig. 3). The value of V_b depends on the PADC material, e.g. it is inversely proportional to the density of crosslinks [2], and on the etching solution (concentration, temperature, etc.) [61]. For a given material and etching procedure, V_b can be taken as constant and the so-called removed layer h, i.e. the thickness of undamaged material removed by the etching, defining the sensitive volume, is simply given by $h = V_b \cdot t$, where t is the etching duration.

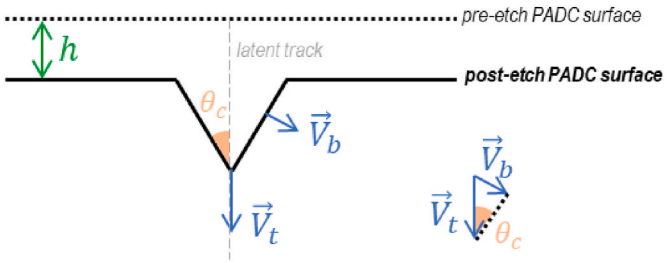


Fig. 3. Sketch showing some geometrical parameters linked to track formation (assuming a constant V_t along the track).

2.2.2. Track etch rate V_t

The track etch rate V_t (μm h^{-1}) is the speed at which the etching solution erodes the damaged material constituting the latent track (Fig. 3). Due to the enhanced chemical reactivity of the damaged material, it results that $V_t > V_b$. V_t is roughly proportional to the LET of the particle generating the track and it follows a "Bragg peak-like" trend along the latent track, i.e. it increases with the decreasing residual range of the particle up to a maximum value reached at the Bragg peak, then it decreases [16]. For many common applications, however, one can consider V_t as constant and equal to its average value along the track. V_t also depends on the material quality and on the chemical etching conditions.

2.2.3. Etch rate ratio V

The etch track ratio V is the ratio between the track etch rate V_t and the bulk etch rate V_b:

$$V = V_t/V_b \tag{1}$$

V is variable along the track, but usually the average value along the track is used.

The value of V is fairly constant among different PADC producers and does not have intra-batch variation. Nevertheless, important decreases in V have been observed with PADC ageing at room temperature and even stronger decreases have been reported because of track fading (see

Section 2.2.8) [13,15]. The reduction in V induced by ageing and fading effects is different for PADC from different manufacturers and is dependent on the specific batch, i.e. it seems to be sensitive to the kind of initiator and to the cure cycle.

The average etch track ratio V along the chemically etched portion of a given track can be estimated from the track geometrical parameters as follows [14]:

$$V = \sqrt{1 + \left(\frac{D}{2h}\right)^2 \left(\frac{2}{1 - \left(\frac{d}{2h}\right)^2}\right)^2} \tag{2}$$

where D and d are the major and minor axes of the ellipse fitting the track opening, respectively, and h is the removed layer.

2.2.4. Critical angle θ_c

The critical angle (or limit angle) θ_c is the threshold angle for track formation: the dip angle θ between the trajectory of the particle which produces a latent track and the PADC surface must be greater than θ_c, i.e. $\theta > \theta_c$ [31]. This is because the track is present in the etched PADC if and only if the erosion along the latent track reaches a depth in the PADC, given by $V_t \cdot t \cdot \sin\theta$ (t = etching time, V_t assumed constant), greater than the one reached by the erosion of the bulk material, equal to $V_b \cdot t$ (Fig. 4), otherwise the track disappears as all the material around it is etched. From this condition, θ_c can be expressed as a function of V_b and V_t [31]:

$$V_t \cdot t \cdot \sin\theta > V_b \cdot t \Rightarrow \frac{V_t}{V_b} > \frac{1}{\sin\theta} \Rightarrow \theta > \theta_c = \arcsin\left(\frac{V_b}{V_t}\right) = \arcsin\left(\frac{1}{V}\right) \tag{3}$$

where the V is the etch rate ratio.

As follows from Eq. (3), V is crucial in determining the critical angle and, thus, the detector sensitivity to radiation. Note that the critical angle is numerically equivalent to one half of the track cone angular aperture (Fig. 3).

2.2.5. Neutron sensitivity

The detector neutron sensitivity, often simply denoted as sensitivity in this work, is defined as the track density (e.g., tracks cm^{-2}) observed in a given neutron field per unit dose equivalent, generally expressed in cm^{-2} mSv^{-1}. It depends on the dosimeter design, including the presence and thickness of neutron converters, i.e. materials coupled with the bare PADC detector to enhance its response (see also Section 2.3.1).

The neutron sensitivity is a function of the neutron energy because of the energy dependence of:

a) the cross-sections for elastic and inelastic neutron interactions in the PADC and in the converter;
b) the range and type of the secondary charged hadrons produced; and
c) the fluence-to-dose conversion coefficients.

It also varies with the irradiation angle, because, for oblique irradiations, less recoil protons satisfy the condition related to the critical angle, Eq. (3), due to the kinematics of scattering.

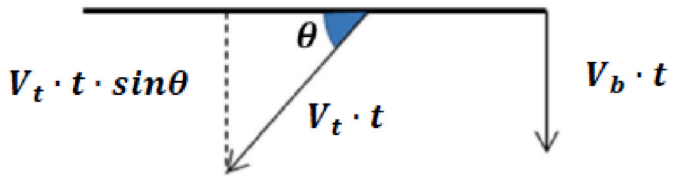


Fig. 4. Sketch for the demonstration of the critical angle formula (assuming a constant V_t). The horizontal line refers to the PADC height just before the etching of the latent tracks begins.

Fig. 5 shows an example of the neutron sensitivity of a PADC dosimetry system as a function of the neutron energy. For dosimeter designs consisting of only fast neutron converters (or no converters) the nominal minimum detectable neutron energy is 100 keV. For lower energies the sensitivity decreases because of: (i) the decreased range of the recoil protons (a 100 keV proton has a range of only ~1 μm in PADC), which results in a decrease in the effective sensitive volume of the detector, and (ii) the fact that tracks with ranges shorter than the removed layer h (typically ~10 μm) may be etched out completely; for neutron energies higher than a few MeV, the sensitivity drops because of: (iii) the reduction in the cross-section for elastic interaction with protons, and (iv) the fact that the recoil proton LET may fall below the detection threshold (for a 20 MeV proton, the LET in PADC is ~3 keV/μm). If the recoil proton LET is lower than the PADC detection threshold, the difference in track and bulk etch ratios is not sufficient to form a visible track.

To illustrate this last point, Fig. 6 shows the simulated LET spectrum of recoil protons behind 2 mm of poly methyl methacrylate (PMMA) due to neutrons from ^{241}AmBe or ^{252}Cf sources, showing that many recoil protons have an LET lower than the typical PADC detection threshold of 10 keV/μm. Nevertheless, with increasing neutron energies, other kinds of high-LET secondaries from inelastic nuclear reactions with carbon and oxygen, most of which have energy threshold above about 10 MeV [72], are produced; their contribution, however, is limited, because the reactions producing them are rarer than elastic scattering on hydrogen [8,9,17].

For dosimeter designs containing a thermal neutron converter and neutron energies between 1 meV and 10 keV, the fluence-to-dose equivalent conversion coefficient $h_p(10)$ is relatively constant, and the neutron sensitivity is essentially determined by the $1/\sqrt{E}$ dependence of the neutron capture cross-section reaction.

Furthermore, the neutron sensitivity is dependent on the PADC material quality, on the chemical etching and on the features of the reading systems adopted for track analysis.

2.2.6. Background

Background is defined as the signal (tracks) detected on an unexposed detector treated using the same procedure as an irradiated detector.

There are different sources of background signal:

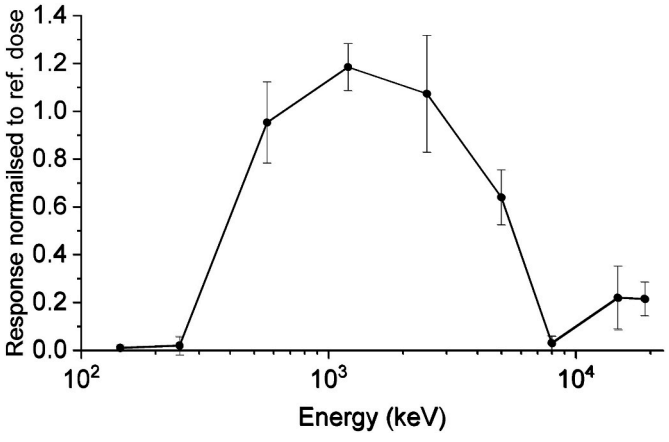


Fig. 5. Neutron response versus energy of the currently adopted neutron dosimeter at PSI, consisting in a 20 × 25 × 15 mm^3 PADC detector coupled to a 2-mm-thick PA12 converter. The irradiated detector undergoes a chemical etching consisting in a bath in a 6.25 N NaOH aqueous solution at 85 °C for 170 min, and the track analysis is performed through a microscope + CCD system. The response is normalized to the response of ^{241}AmBe. Data obtained by irradiation with monoenergetic neutron beams at PTB in 2010. Redraw based on data from Ref. [54].

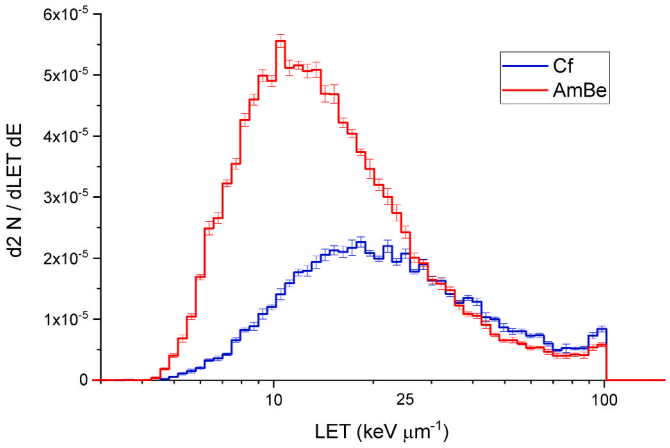


Fig. 6. Recoil protons *versus* LET in water generated by neutrons interacting with a 2 mm PMMA converter simulated for the neutron spectrum ^{241}AmBe and ^{252}Cf sources using the FLUKA code. Reproduced from Ref. [77].

(a) **natural background (cosmic rays or radon)**: cosmic rays (not limited to the neutron component) can create tracks in PADC, which must be experimentally quantified if needed. α-particles from radon and its daughters can normally be easily discriminated based on track parameters, since they are generally larger and more elliptical than the neutron-induced tracks from recoil protons (due to their higher LET) (compare Fig. 7a and b).

(b) **scratches and dust**: the signal associated with scratches (Fig. 7c) and dust are easy to limit or even suppress completely by the careful manipulation of the dosimeters (see Section 3.1.4) and through solutions such as pre-etch or film to protect the dosimeter surface.

(c) **intrinsic background:** this is caused by internal plastic defects/bubbles inside the material, related to the way the PADC is produced, that lead to a signal recognized as neutron tracks through the analysis system (see Fig. 7d and e).

2.2.7. *Detection limit (DL)*

The detection limit (DL) is "the smallest true value of the measurand which ensures a specified probability of being detectable by the measurement procedure", as defined by the ISO 11929–1:2019 [46], which also provides a general guideline to estimate it. If the background is Poisson distributed and a confidence level of 95% is chosen, a practical estimate of the detection limit is given by Ref. [21]:

$$DL = \frac{C}{A}\left(4.65\sqrt{B} + 2.71\right) \tag{4}$$

where B is the average background in terms of tracks, A is the detector area and C is the calibration factor to convert from track density to dose.

Due to the intrinsic complexity of neutron dosimetry and the main

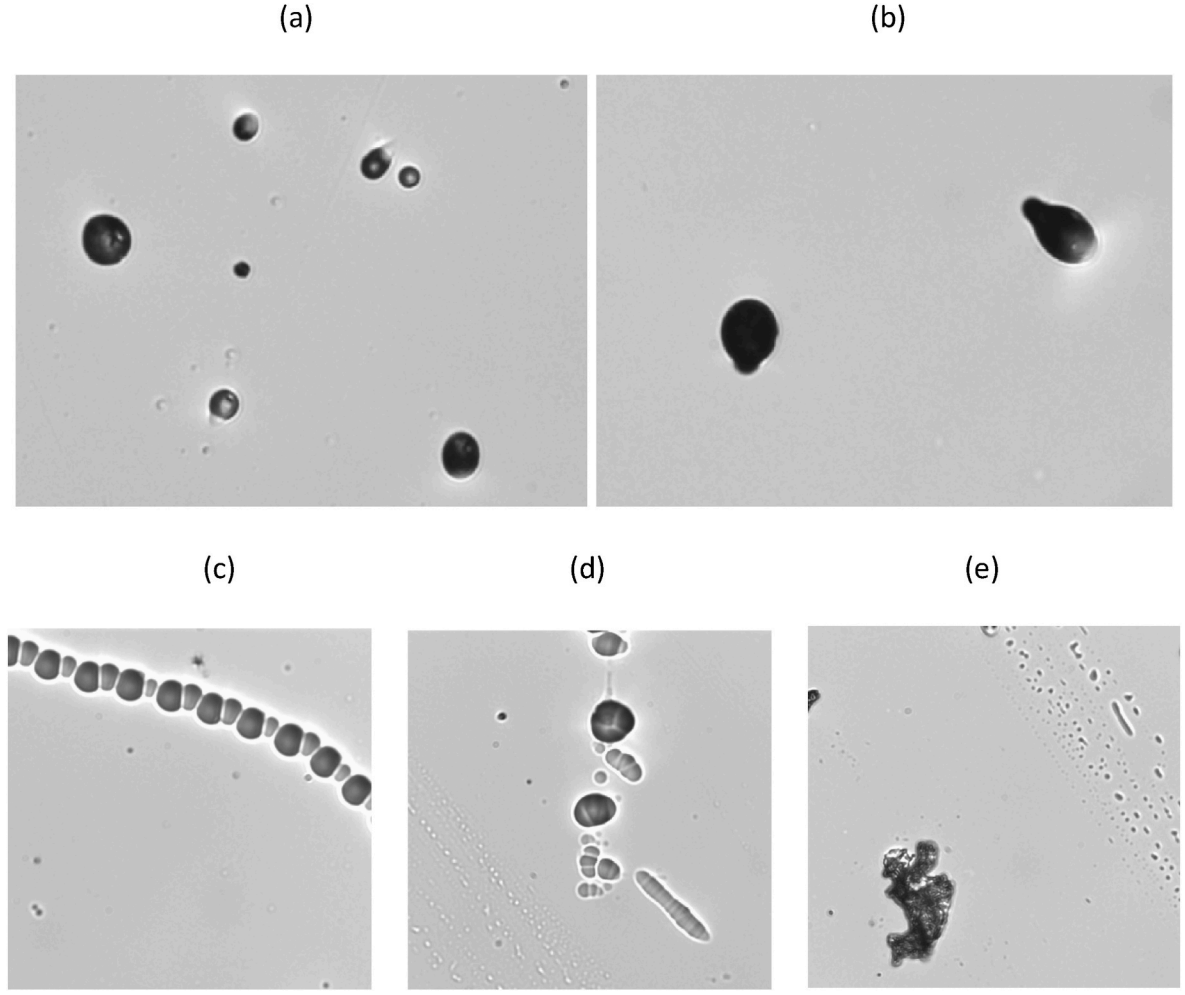


Fig. 7. Comparison between (a) neutron-induced tracks (from a ^{252}Cf source) and (b) alpha particle tracks due to radon/radon daughters (generated on a test PADC exposed to air), for a fixed etching procedure and microscope magnification. (c) Scratch. (d–e) Examples of plastic defects contributing to the intrinsic background: (d) bubbles, (e) amorphous defect.

issues linked to PADC quality (see Section 1), the DL of PADC-based neutron personal dosimeters typically is 0.1 − 0.2 mSv [80].

2.2.8. Fading and ageing phenomena

The terms *fading* and *ageing* are used to express a decrease in the PADC neutron sensitivity with time. Fading refers to the decrease in sensitivity due to latent track repair ("damage healing" or "annealing") during and after the use of the detector. Ageing expresses a worsening of the detector material quality, resulting in a loss of sensitivity, and it is observed when the detectors are stored for some time before use. These effects strongly depend on time and storage temperature, becoming important on a timescale of months and for temperatures at or above room temperature.

Previous studies showed that the ageing/fading effect is strongly dependent on temperature [13,15]. Detectors stored in freezer (−18 °C) prior to and after irradiation showed low or no ageing and fading, while detectors kept at environmental temperature were heavily affected. Fading and ageing cause a reduction in V and consequently a reduction in the PADC detection sensitivity. Ageing mitigation by storage at low temperature is impossible during the on-field exposure period (typically 1 or 3 months), so the impact of ageing is almost impossible to foresee. Fading appears only during the on-field exposure (i.e., once tracks are created) and there is no possibility of controlling it. Between the two effects, fading is more severe because, under the same temperature conditions, faded tracks on unaged PADCs exhibit a lower V when compared to non-faded tracks on aged PADCs.

The reduction rate in V is highly dependent on the quality of the material and can be different among material from different manufacturers and, even from the same produces, intra-batch variation is reported, based on the experience of the IMSs participating in this project. Whereas these effects can be compensated for PADCs exposed to radon, based on the fact that the alpha particles emitted are monoenergetic, for neutron detection the compensation is not possible; thus, this is a critical aspect. Nevertheless, the shorter irradiation period (1–3 months) of PADC used for neutron dosimetry compared to the typical period of radon exposure measurements (6–12 months) helps mitigate the impact of these effects.

Protecting PADC from UV light limits these effects too [36,37].

2.3. Practical considerations

Various practical aspects regarding converters, etching procedure, reader and track analysis must be considered when using PADCs for neutron dosimetry.

2.3.1. Neutron converters

Converters are materials placed in front of the PADC to convert neutron radiation into charged secondaries, enhancing the signal registered [43]. The type of converter used depends on the radiation to be detected:

a) fast neutron converters typically consist of hydrogenous materials in which fast neutrons (i.e., neutrons with energy above 100 keV) induce secondary charged particles, mainly recoil protons, which can produce etchable and detectable tracks in PADC;
b) thermal neutron converters refer to materials containing nuclides with large thermal neutron capture cross section (such as ^{6}Li, ^{10}B or ^{14}N), which capture neutrons to generate charged secondary particles (e.g., alpha particles, tritons, etc., plus residual nuclei) with an energy determined by the reaction Q-value [51] and which can produce tracks in the detector. Note that these converters also detect some epithermal neutrons, but with a sensitivity scaling with energy as $\sim 1/\sqrt{E}$: hence, the epithermal signal is usually neglected. If necessary, however, the epithermal signal can be accounted for and discriminated from the thermal one by using a Cd filter placed in front of the converter film to block the thermal neutrons. Similarly, reactions with fast neutrons are generally neglected due to the low value of the neutron capture cross section for fast neutron energies.

Some authors reserve the term "converter" for thermal neutron converters, whereas fast neutron converters are called "radiators" [64].

Notice that if no fast neutron converters are used, secondary particles are generated in the removed layer only. Adding a fast neutron converter can increase the neutron sensitivity to high neutron energies. As shown in Fig. 8, the build-up layer until charge-particle equilibrium (CPE) is achieved varies with the neutron energy; for the ^{241}AmBe and ^{252}Cf neutron spectra, ∼1 mm of polyethylene is required to achieve CPE. Therefore, the neutron sensitivity can be increased by adding a converter with sufficient thickness to achieve CPE. Furthermore, for the same thickness, the higher the hydrogen content of the converter, the higher fluence of recoil protons (Fig. 9). Some IMSs exploit the PADC volume itself as a converter by looking at tracks produced on the "rear" face of the detector, the entire detector serving as a neutron converter [80].

Finally, note that the choice of the reactive isotope content in a thermal neutron converter is important, because too many reactions may saturate the detector.

2.3.2. Etching

The objective of the post-exposure etching of the PADC detectors is to create tracks that are large enough to be observed under the optical microscope (i.e., with diameter greater than a few microns, depending on the system magnification) and fix them in the PADC material. The track size will result from the combination of material, etchant, etchant temperature and etching time, in addition to the particle LET.

Fig. 10 illustrates the relationship between the etching solution temperature for a 6.25 N NaOH aqueous solution and the bulk etch rate.

For a given material and etchant, the optimum etching time can be established by measuring the curve of sensitivity versus time. As already mentioned, the etchant typically is an aqueous solution of NaOH or KOH, which are strong bases which fully dissociate in water to produce hydroxide ions useful for the etching process. For a given solution temperature and molarity, KOH shows a greater V_b than NaOH, so that a shorter etching time is required to remove the same PADC layer [4,61].

In the electrochemical etching (ECE) process, a preliminary stage,

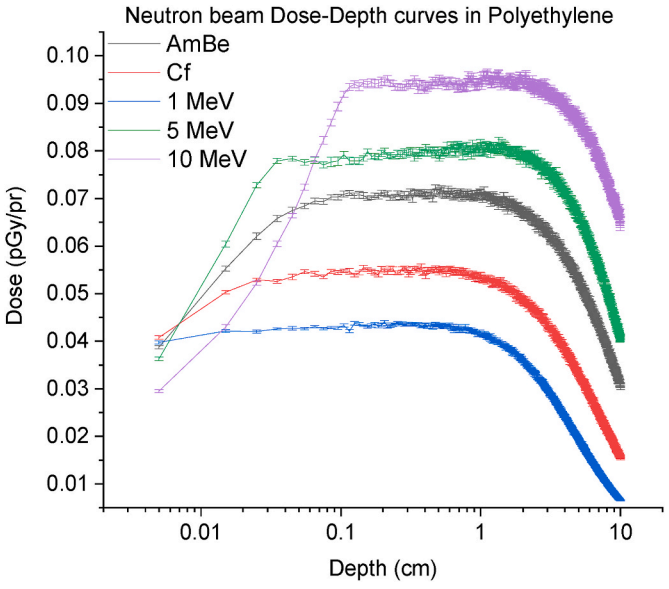


Fig. 8. Dose-depth curves in polyethylene for mono-energetic neutron beams with different energies, as well as for the neutron spectrum from ^{241}AmBe and ^{252}Cf sources, simulated using the FLUKA code. Reproduced from Ref. [77].

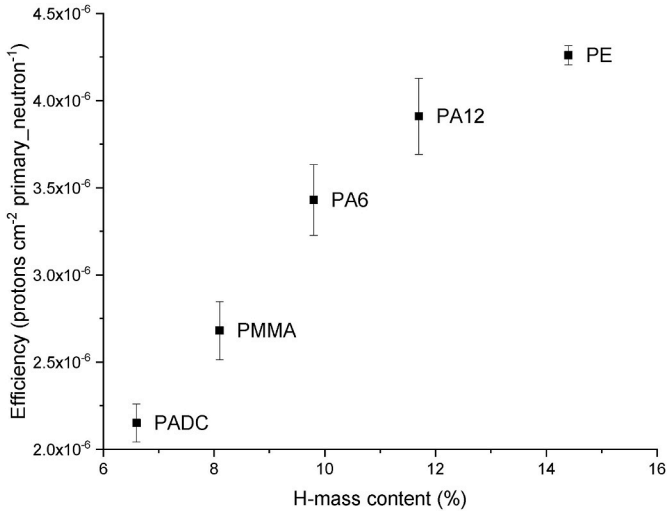


Fig. 9. Conversion efficiencies of several materials, typically used as converters, as function of the hydrogen mass content in the material composition. Reproduced from Ref. [77].

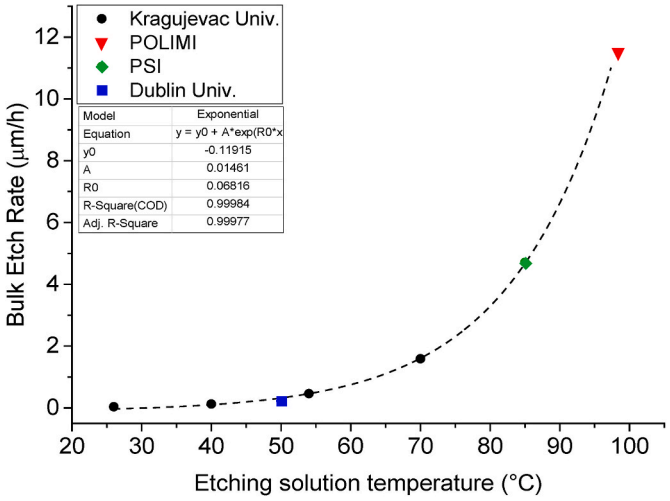


Fig. 10. Trend of the bulk etch rate VS temperature for a 6.25 N NaOH aqueous solution. Values refers to current parameters used at POLIMI and at PSI, additional values were obtained from other research centres. Reproduced from Ref. [76].

with no electric field applied, is required to initiate the etched pits [26,38]. This post-use stage is sometimes called a "pre-etching" and should be distinguished from treatments intended to remove surface defects before the PADC is issued, confusingly also called "pre-etching". The second stage of the process involves application of an alternating electric field. Because the impedance is lower where the PADC is thinner, the current preferentially flows through the etched pits. This causes damage which, with the right parameters, can be visible to the naked eye and counted under low magnification devices, such as microfiche readers or slide projectors.

In ECE, there are thus more parameters to be optimized.

- Choice of etchant. As for ordinary chemical etching, this is usually an aqueous solution of KOH or NaOH, with similar strength, e.g. 5 N.
- The preliminary etching temperature and duration. This can be "short and hot", e.g. 1 h at 70 °C, or "long and warm", e.g. ~12 h at 40 °C.
- ECE field strength and frequency. Field strengths between 20 and 25 keV cm^{-1} and frequencies between 50 Hz and 2 kHz have been investigated [35,79].

As for ordinary chemical etching, optimization must consider the detection limit, the energy detection threshold and the overall energy dependence of response. Further, with a two- (or more) stage process, there will also be several practical constraints.

Note that, whereas chemical etching is performed in common thermostatic tanks, electrochemical etching requires a dedicated setup with a more complex design (ECE cell) [26].

As mentioned above, a pre-etching (PE) to remove a few tens of µm can be performed by the IMSs to erase surface defects before sending the dosimeters to the customers. This treatment is important if the PADC supplier does not protect the surface of the plastic after its production (with a film, for instance), which can result in damage (scratches) and accumulation of radon-related tracks. This PE can also adjust the PADC thickness to the size of the holder. However, PE does not drastically modify the global metrological performances of the material whereas it significantly increases the cost of the dosimetry service (in terms of labour and resources, it is similar to the post-issue etching process, which is itself one of the main contributors to overall cost), and it means for the IMS one more step before sending the dosimeters to the customers; hence, although the pre-etching process has been used for research applications, it is not widely used for individual monitoring where it is avoided as far as possible by using other, cheaper means to protect the PADC surface.

2.3.3. *Imaging system*

The design of most systems is essentially similar, consisting of a transmission optical microscope coupled to a digital camera. PADC detectors are placed on a motorized X–Y plate, for the automatic scanning of their surface. The main parameters characterising a particular imaging system are the illumination, effective resolution, field size and total scanned area.

The illumination system is usually the standard one commonly used in optical microscopes. The choice of the light is not critical, since PADC is transparent in the whole visible light spectrum. LED illuminators emitting in a specific colour are sometimes used [57,82]. A light condenser can be used to obtain a more uniform grey level (i.e., pixel brightness level) over the field of view of the microscope, particularly in case of high magnification. The stability of the light source is critical because it can induce a drift in the background and tracks grey levels. The variation can be compensated for by changing the camera gain. Some readers have a manual or automatic procedure to keep the grey level constant by tuning the gain.

The effective resolution (i.e., the system ability of resolving details) depends on the optical tube, which is composed by an objective and an eyepiece. The latter can be substituted by a pinhole that has the advantage of increasing the focus depth, allowing autofocus routines less sensitive to sample-objective distance variations to be implemented. The pinhole can be used with objectives with magnification 4 × or 10 × , while for higher magnification it results in a lack in grey level uniformity.

The total magnification is more conveniently expressed in terms of the area corresponding to the pixel dimension, indicating the spatial resolution of the system. The pixel dimension along with the camera resolution (i.e., the number of distinct pixel in each dimension, typically 1280 × 960 pixels) define the field size, namely the area of each frame. If a simple counting of chemically etched tracks generated by alpha particles is needed, magnification leading to 1× 1 μm^2 per pixel (field size around 1 mm^2) is generally enough. In the case of neutron dosimetry, most of the tracks are due to recoil protons and are considerably smaller (area of tens μm^2, usually), and the pixel dimension shall be around 0.5 × 0.5 μm^2 to obtain a suitable accuracy with which some track

parameters, such as track lateral dimensions and area, can be measured. The finer magnification has some impacts: (i) the focus depth is lower and the autofocus routines must be reproducible with a tolerance of a few μm – the criticality of the autofocus impacts on the overall reading repeatability (ii) the filed size is smaller and number of frames to scan becomes considerably higher, increasing the reading time. The best way to measure the field size and the effective pixel dimension is by using a micro-ruler with a resolution of at least 10 μm.

Finally, the total scanned area is defined as the area of the total number of frames that are processed during the scanning. The scanned area is defined to get a statistically meaningful number of tracks. It can be defined *a priori* or by stopping the acquisition once enough tracks to achieve the desired uncertainty are detected. It should be noted, however, that the final uncertainty on the calculated dose is affected by additional sources of uncertainty (i.e., detector background signal, reference exposure, etc.), and therefore the benefit of increasing the total scanned area becomes negligible above a certain limit. Monte-Carlo simulations and measurements with very sensitive Fluorescent Nuclear Track Detectors (FNTDs) show that track densities of the order of 1000–2000 tracks cm^{-2} mSv^{-1} can be achieved behind 2 mm of polyethylene in the case of ^{241}AmBe and ^{252}Cf neutron spectra [75]. In practice, PADCs generally have a lower sensitivity for the same spectra, owing to the material and analysis algorithm, which is around 300–1000 tracks cm^{-2} mSv^{-1}, depending on the specific system [80]. Therefore, a dose of 0.1 mSv may correspond to few tens of tracks per cm^{-2}, indicating that a scanned detector area of the order of 1.0 cm^2 would be required to achieve reasonable statistics.

As the dimension of the detector is few cm^2, the neutron field can be considered in most cases uniform over the PADC area, meaning that every single frame can be seen as a detector observing a number of events extracted from the same Poisson distribution. Thus, statistical tests can be applied to evaluate the plausibility of a certain distribution of tracks among the frames to indicate the possible presence of scratches or defects mimicking tracks.

2.3.4. Track analysis

The acquired images are analysed with dedicated software that implements a track analysis algorithm. Not only is this software as important as the hardware for the final performance of the neutron dosimetry system, but the development of the track analysis algorithm can be equally time-consuming as the hardware design and construction.

Different analysis software implement different features, which are also related to the dosimetric approach used for the dose estimation (see Section 2.4). In general, however, the main features include the identification of potential tracks, measurement of track features, and discrimination or filtering of background tracks. Commercial software may already be optimized, but usually users have the option to change some parameters (e.g., minimum track size, grey level threshold).

The first step in the analysis consists in identifying the tracks present in each frame. This is done in two steps: the system identifies the objects (possible tracks) present in each frame as compact region (group) of pixels with a grey level above the pre-set threshold value and it calculates some geometric and optical parameters associated to each object. It then selects the objects corresponding to "real" tracks based on the object parameters.

Since hundreds of images per detector may be acquired for statistical purposes to achieve the required detection limit, the images corresponding to the frames are typically not stored in commercial dosimetry systems to save storage space. Instead, only the object parameters are stored. The parameters recorded by each system differ, but the most general are the area, perimeter, mean grey level inside the track, major and minor axes of the ellipse that best approximates the shape of the track ("fitting ellipse") and a (system-dependent) parameter indicating the goodness of the track fit. Besides discriminating real tracks from background, these parameters are useful for refining the dosimetry approach (see Sections 2.4.1–2.4.2). Moreover, there is a correlation between the geometry of the track and the transmitted light intensity (brightness) – for tracks with larger depth, the track core is more intense because of lesser attenuation of the transmitted light, and iso-density contours of the grey levels can be employed for the separation of partially overlapping tracks and to extract information on 3D shape of the tracks [22].

It is worth mentioning that the dimensional parameters can be compared among different reading systems, whereas the grey level is a system-dependent parameter because it is strongly affected by the illumination conditions. Using 8-bit CCD cameras, the grey level ranges for 0 to 255. The background grey level is set to a system-dependent reference level that can range from 180 to 230. As the track grey level is measured referring to the background grey level, it is clear this parameter cannot be used as an inter-systems comparison.

As already mentioned, from the parameters measured above, and potentially others, different criteria can be applied to discriminate between tracks generated by the neutron and background tracks. For example, shape-based filtering can be applied to count only tracks with pre-selected value of roundness: particle tracks usually have circular and almost elliptical pits, whereas defects, scratches, and artefacts are of any sizes and shapes (however, low-contrast elongated tracks produced by particles with shallow angle of incidence would be excluded from the signal by this approach).

It should be noted that any criteria aimed at reducing the number of background tracks will, in general, also result in a reduction of neutron-induced tracks and, consequently, of the sensitivity of the overall measuring system; a trade-off should therefore be achieved between background track density and sensitivity, searching for the maximum signal-to-noise ratio.

The possible routine to filter out the spurious signal in track detectors are various, from the use of a simple set of filters on the geometrical parameters (mainly track dimensions) or optical parameters (especially the mean grey level) [6,22,50,86], to methods based on the comparison with a database of "reference" tracks [62] to more recent and sophisticated statistical techniques such as the Principal Component Analysis (PCA) [74].

From the measurement of the track minor and major axes it is also possible to determine the particle LET, which is required in some dosimetric approaches (see Section 2.4.2). This kind of analysis requires a high resolution to measure the track parameter with a level of accuracy higher than that required for simple track counting, hence a greater objective magnification is required (e.g., 20 ×).

In conclusion, it is important to mention that the settings of the reading system depend not only on the final performance required, but also on the constraints specific to each IMS (quantity of dosimeters to be analysed for instance): image resolution, image sampling and image analysis may be set considering this need of a higher productivity.

2.4. Dosimetry approaches

There are three main methods to calculate the neutron dose from fast neutrons: (i) based on the track density (after a track filtering based on track parameters) with the application of appropriate calibration and correction factors, (ii) based on the analysis of track morphology to obtain an LET spectrum and calculate the resultant neutron dose, and (iii) based on bulk properties (e.g., light scattering or light transmittance). Furthermore, the information from PADC behind a fast neutron converter and a thermal neutron converter can be combined to improve the energy response of the dosimetry system.

2.4.1. Track density

Neutron dosimetry using PADC, both for fast and slow neutrons, is typically based on the evaluation of the track density (e.g. tracks cm^{-2}) behind the neutron converter and assuming a linear relationship between the track density and the personal dose equivalent $H_p(10)$:

$$H^M = N \cdot M \cdot \prod k_i \tag{5}$$

where here we are using the symbols traditionally used in the ISO 21909–1:2015 [48]: H^M is the indicated value for the PADC taken as an estimation of the personal dose equivalent, N is the calibration factor, M is the background-corrected PADC reading (track density), and k_i are correction coefficients to account for fading, energy dependence or other influencing factors.

The calibration factor N is determined by irradiating calibration dosimeters from the same batch in reference conditions and calculating the ratio between the reference quantity value H^0 and the average reading M of these dosimeters:

$$N = \frac{H^0}{M} \tag{6}$$

Neutron reference fields and calibration procedures are described in the ISO 8529 series [44,45,47], but some added specificities could be given by national regulations.

If necessary, control (unirradiated) dosimeters can be used to estimate the background signal and subtract it from the estimation of the neutron personal dose equivalent. These control dosimeters may indicate an increase in the background track density due to other sources, such as material ageing, as well as track caused by cosmic rays and radon decay products.

Both calibration and control detectors should be from the same batch for the specific period of exposure. If needed, fading can be corrected for by characterizing it directly or irradiating the calibration dosimeters at the beginning or middle of the monitoring interval.

Sometimes correction factors based on track size are applied to the simple track density to extend the validity of the calibration factor to a wider range of neutron energies and neutron impinging angles, which can be helpful to comply with the requirements of ISO 21909–1:2021 [48,58]. Corrections for track overlap that occurs at high doses have also been proposed [85,88].

If a combination of converters is used to extend the application energy range of the dosimeter (e.g., polyethylene for fast neutrons and a converter containing ^{6}Li for thermal neutrons), then an algorithm must be used to combine the information from the various PADC portions [53]. The main limitation is that the spectral sensitivity coefficients used in the algorithm, especially the ones related to thermal neutrons, show a strong dependence on the energy of the neutron fields [5,30,54]. The thermal neutron component, in particular, can vary significantly with the materials around the dosimeter (person's body, walls and other neutron scattering components). Therefore, accurate dose estimations rely on the quite restrictive assumption that neutron fields, where dosimeters (and people) were exposed, must have a very similar spectrum to the one used for the calibration of dosimeters. Besides, in non-isotropic fields, such coefficients are also known to vary with the angles of exposure [80], hence introducing additional uncertainty.

2.4.2. LET spectrometry

PADCs can be used to determine the LET spectrum of a charged particle radiation field, as already commonly done in the field of space dosimetry [7]. Since the neutron dose is deposited in the materials by secondary charged particles, the LET spectrum obtained using PADCs can be used to estimate the absorbed dose in the PADC due to neutrons. As not all secondary particles are registered by the PADC (see Sections 2.2.4–2.2.5), the efficiency of the detector is < 1 and a proper calibration against other quantities of interest, e.g. operational quantity $H_p(10)$, must be established.

The LET spectrum can be obtained by calculating the LET of the particles generating the tracks. For a given particle associated to a chemically etched track the registered LET can be obtained through a relation between V and LET, where V can be calculated using the track opening minor and major axes, as well as the removed layer h, through Eq. (2). The relation between V and LET of the particle (calibration) is usually expressed analytically with a polynomial function that fits the experimental data obtained by irradiation of the detector's material in known heavy ion beams [16,25,65].

The dose D (in mGy) and the dose equivalent H (in mSv) respectively can be then estimated as [16]:

$$D = \frac{1}{\rho A} \cdot 1.602 \cdot 10^{-6} \cdot \sum_{i=1}^{n} \frac{\overline{LET}_i}{\cos \alpha_i} \tag{7}$$

$$H = \frac{1}{\rho A} \cdot 1.602 \cdot 10^{-6} \cdot \sum_{i=1}^{n} \frac{\overline{LET}_i}{\cos \alpha_i} Q_i \tag{8}$$

where $\overline{LET}_i$ is the mean LET along the i-th track limited to the track portion affected by the etching in keV μm^{-1}, A indicates the detector (scanned) area in cm^2, α_i denotes the particle impinging angle with respect to the PADC surface normal (computed starting from the track opening axes as illustrated in Ref. [16]), ρ is the PADC density, Q_i is the ICRP quality factor and the index i runs on all the n etchable and detectable tracks.

For practical purposes, the quantity of interest is the dose equivalent H in PADC, which can be related to the operational dosimetric quantities of interest (e.g., $H_p(10)$ for personal dosimetry) through proper calibrations. This can be done by irradiating the dosimeters in various neutron fields, covering the energy and angle ranges of practical interest, and evaluating the ratio between the calculated dose and the reference quantity, $H_p(10)$ or $H^*(10)$, to set a numerical factor which, multiplied (or divided) by the calculated dose, allows to obtain the desired dosimetric quantity within a certain confidence interval.

This approach leads to a response with a reduced energy and angle dependence compared to the one based on the simple track density [8,9,17].

Although this dosimetry approach is mainly applied in research at present, there are commercial readers adopting it, like the *Politrack*® system (Mi.am Srl, Piacenza, Italy) [57].

2.4.3. Bulk properties

Bulk properties, such as the light scattered by the etched tracks and the light transmission, can also be used for neutron dosimetry.

The total light scattered by the PADCs when illuminated on the side was used by the *Autoscan 60* system (Thermo Electron Corporation, Beenham, UK) for neutron doses between 15 mSv and 100 mSv [29]. For higher doses (above ~100 mSv), the track density increases and tracks start to overlap (Fig. 11), and the procedures based on the recognition and counting of individual tracks become not feasible. In this case, however, a calibration curve can still be established between the overall light transmission or absorbance in the etched detector and the neutron dose [10]. This can be accomplished using a dedicated spectrophotometer, but it is also practical to use the microscope images obtained with the equipment used in the routine dosimetry [73].

The calibration curve consists in a curve of sample absorbance, calculated based on the mean grey level of the microscope images, versus dose. In the 100 mSv–1 Sv dose region, a generic dose response curve can be used for dosimetry, as long as the response of the same batch to 1 Sv is determined for the normalization. Above 1 Sv, however, the uncertainty becomes large and a detailed response curve for the same batch must be determined [73].

2.4.4. Calibration procedures

Neutron personal dosimeters are commonly calibrated by use of ^{241}AmBe or ^{252}Cf neutron sources, with irradiations carried out in accordance with ISO 8529 [44,45,47] (see Section 2.4.1 above). Of these, ^{241}AmBe has the much longer half-life and such sources will require replacing less frequently. As part of their routine practice, metrology laboratories should periodically check to make sure the ingrowth of decay products is not affecting the source fluence energy

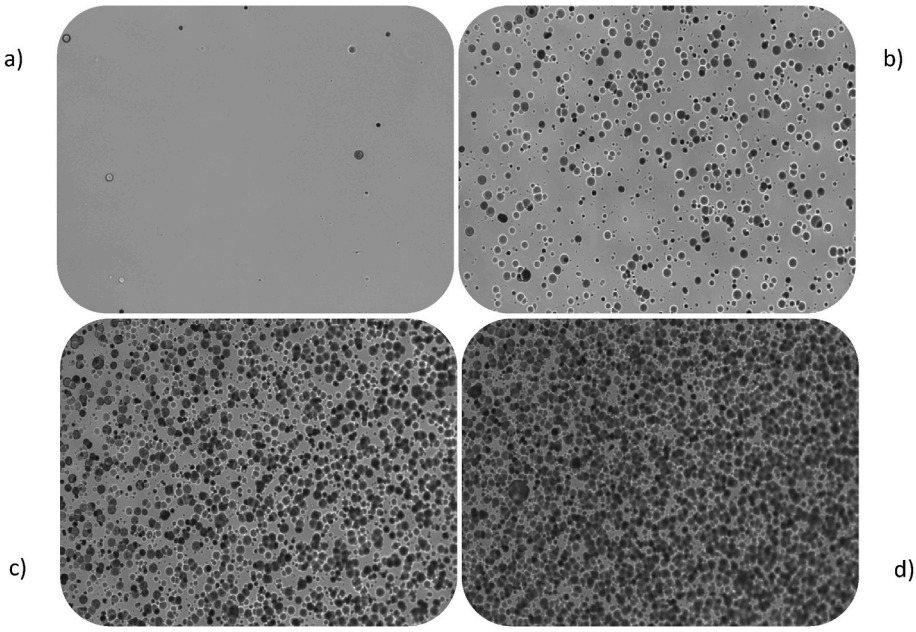


Fig. 11. Track patterns on an area of about 620 × 470 μm^2 for detectors irradiated with neutrons from a ^{252}Cf source: (a) 10 mSv, (b) 0.5 Sv, (c) 2.5, and (d) 5 Sv. Reproduced from Ref. [76].

spectrum; this is more likely to be an issue with the shorter-lived ^{252}Cf. Calibrations involving monoenergetic neutron fields are less frequent, mainly due to the high cost required to access facilities where such fields are produced.

Because neutron workplace spectra are generally different from the above calibration spectra, it is recommended that a measurement survey of the typical spectra found in the clients' workplaces be made. The survey should address the energy and angle dependence. Exposure situations will often be approximated by the following:

- A-P (antero-posterior) – the neutron radiation comes mainly from in front of the wearer.
- ISO (ROT) (rotationally isotropic) – the radiation comes approximately uniformly from all directions about a vertical axis through the wearer. If a worker moves about in the radiation field, this can render the distribution effectively isotropic.

Situations where spherical isotropy is appropriate are uncommon; and if the predominant direction of incidence is from behind the worker, the dosimeter should be worn on the back. In a more general way, some methodologies to characterize the workplace field are described in the ISO 21909–2:2021 [49]. Once the spectral energy and angle distribution is known, this can be taken into account by Ref. [33]:

a) Establishing a computational model (e.g., MCNP) based on the dosimeter type test results for energy and angle dependence of the response, including monoenergetic, thermal and radionuclide source data.
b) Calculating a correction factor for each of the workplaces covered by the dosimetry service, using knowledge of the neutron energy and angle distributions in those workplaces.
c) Calculating the average correction factor over all workplaces.
d) Applying a correction factor to all reported dosimeter results. Depending on the requirements, the correction factor can either be the overall average, or the workplace-specific value.

Nevertheless, establishing a coefficient from characterizations of different workplaces is not common practice for PADC. This method is very difficult to implement, based on the assumption that work situations do not change, whereas the principle of PADC is rather based on obtaining an energy response considered sufficiently flat to obtain reliable dosimetry results without prior knowledge of the workplaces.

2.5. Technical requirements

The International Standardisation Organization (ISO) Technical Committee (TC) 85/SC 2 Working Group 19 dealing with "Individual monitoring for external radiations" has revised the international recommendations for passive neutron dosimetry systems. The current version of the ISO standard ISO 21909 is supposed to be constraining enough to ensure that any neutron dosimetry system fulfilling the outlined criteria would be reliable in most of the usual work situations in terms of dose level and neutron spectra. This ISO standard is split into two parts: Part 1 [48] provides performance and test requirements for measurement of personal dose equivalent $H_p(10)$, for neutrons ranging from thermal energy to approximately 20 MeV (this upper limit being considered as covering most of the situations of occupational exposures). Part 2 deals with systems which do not meet the criteria regarding energy and directional dependence of response described in part 1 but are able to give consistent and reliable dosimetry at selected workplaces by applying corrections when needed.

3. Best practices

As any dosimetry technique, PADC neutron dosimetry relies on a careful choice of parameters and a rigorous quality assurance program. In this section we summarize the best practices to achieve a good quality in this field based on the experience of the authors (Table 1).

3.1. PADC material and handling

Choice of material and material quality assurance tests are important to achieve the performance requirements in PADC neutron dosimetry. A clear documentation of the acceptance test results and a regular dialogue with the manufacturer can help improve the material quality and consistency over time.

Table 1
Summary of quality assurance tests typically used in PADC dosimetry.

Test	Objective	Parameters monitored
Material acceptance test	Control the quality and consistency of the PADC material	• Mean and standard deviation of the background per sheet/batch • Mean and standard deviation of the sensitivity per sheet/batch
Control of the etching procedure	Control the removed layer/Achieve reproducible track size	• Dose of irradiated detectors • Chemicals • Temperature and timing • Molarity • Field strength (for ECE)
Blind tests	Control of the entire dosimetry process	• Dose of irradiated detectors in reference conditions
Intercomparisons	Control of the entire dosimetry process and some dosimetric properties	• Dose of irradiated detectors in various conditions

3.1.1. Choice of material

Except for custom-built systems, in which the IMS has full control of the reader and algorithm parameters, choice of the material normally implies choice of the entire dosimetry system (material, etching, reader, analysis algorithm), because of compatibility issues between material, reader and analysis algorithm. Compatibility issues also include different detector sizes and thickness, as well as the system for detector identification.

It is important to properly choose the PADC material with respect to the planned application. Characteristics that are dependent on the material include the detection neutron energy range, background, sensitivity (including inter-batch and intra-batch dispersion, as well as long-term stability), and sensitivity to environmental effects. These properties may differ considerably among the various manufacturers.

The minimum detector size required will depend on the sensitivity (tracks cm^{-2} mSv^{-1}). The detector should be sufficiently large to provide the required statistics and detection limit with the available scanned area.

3.1.2. Material acceptance test

Because the quality of the material may vary significantly not only from batch to batch, but also from sheet to sheet, even for material from the same manufacturer, it is essential for the quality of the final result that an acceptance test is performed for each sheet. Since this analysis is destructive, a balance must be made between obtaining a representative sample of the batch and still having sufficient detectors for the routine use.

Typically, the IMSs use 10% of the detectors in each batch for the acceptance test, 5% for check of the batch sensitivity and 5% for check of the background signal. For the acceptance test, detectors can be irradiated with a known dose and, along with the unirradiated detectors, etched and evaluated. This provides an estimate of the sensitivity and background signal of each sheet and, combined, of each batch. As the variability of the background is also important, it is recommended to have different criteria: one for the mean value of the intrinsic background and one for its standard deviation.

Fig. 12 shows an example of the results of an acceptance test for four batches for both unirradiated and irradiated detectors. The figure shows the track density for five unirradiated and five irradiated detectors (3 mSv, ^{241}AmBe) for each sheet. Fig. 12a shows that some sheets present an unusual number of unirradiated detectors with a high track density (>50 tracks cm^{-2}). Fig. 12b shows that some sheets have unusually low sensitivity (<150 tracks cm^{-2} mSv^{-1}, corresponding to a track density <450 tracks cm^{-2}). Such sort of data must be analysed to determine if

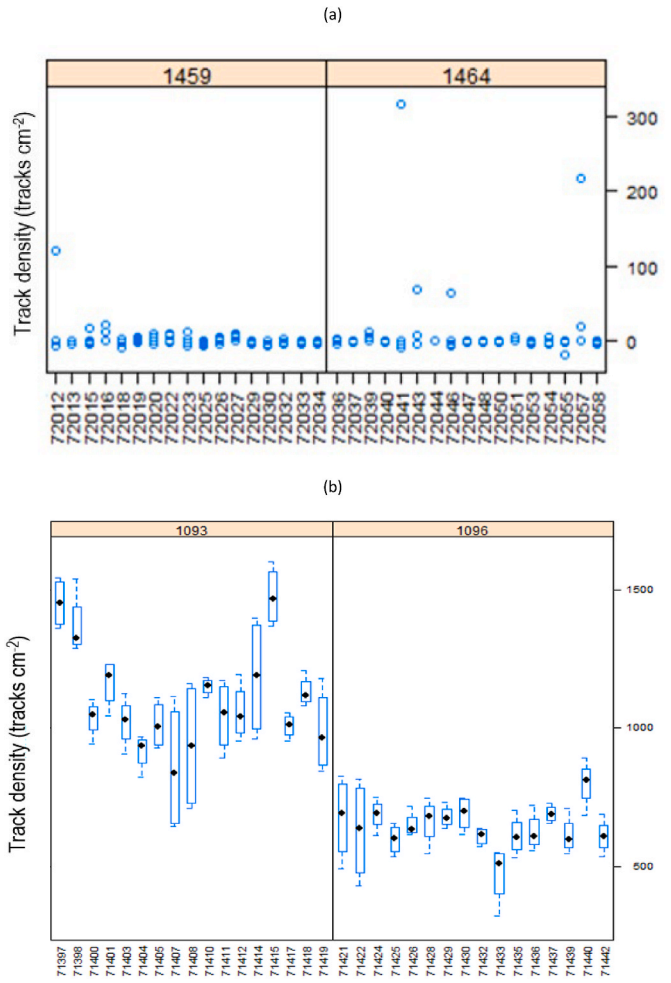


Fig. 12. Example of acceptance test results for (a) unirradiated detectors and (b) irradiated detectors for various batches (four-digit number) and sheets (five-digit numbers).

the batches and sheets can achieve the dosimetric requirements, for example, in terms of reproducibility and minimum detectable dose.

If a batch shows sensitivity that can be clearly separated into two groups, due to the influence of some parameters in the material production (e.g., curing at two different temperatures), the batch should be divided into two separate (sub-)batches.

Furthermore, we recommend using only a single batch for each monitoring period, if possible, since other properties such as material ageing and signal fading can differ from batch to batch. For IMSs that have a number of workers to be monitored such as one batch could not be sufficient per monitoring period, then batches with similar characteristics in terms of background (mean and standard deviation) and sensitivity (at least) should be used when possible, otherwise the difference in sensitivity of the batches employed must be considered when reading the dosimeter and calculating the dose (i.e., referring to a different calibration factor and/or detection limit).

Although the acceptance criteria are defined by each laboratory to guarantee that they satisfy the requirements for the dosimetry service, we recommend the preparation of an acceptance test report that can be used to clearly demonstrate the compliance or non-compliance of the sheet/batch characteristics with the stated criteria. This is useful when discussing the results with the manufacturers.

3.1.3. Material storage

To reduce or suppress changes in sensitivity and detector response with time of storage prior to the irradiation (ageing), the material should

be stored under a low temperature and without air access. Practically, these conditions are met by storing the detector sheets in freezers inside radon-proof bags (e.g., heat-sealed mylar bags), avoiding rubbing the sheets together while manipulating them to prevent scratches. Sometimes PADC sheets are already sold wrapped in plastic films to protect them from damages due to manipulation (like scratches) and from radon contamination. Alternatively, sheets can be stored under nitrogen to prevent both oxidation and radon contamination.

To minimize the effect of fading, the detectors should be etched soon after the end of the monitoring period and stored in freezer and protected from air before undergoing the etching.

One should aim at having a constant storage time between receipt of the PADC from the manufacturer and issuing it. This is not always possible, considering both economies of supply scale and periodic variations in issue demand, but avoiding excessively long or excessively short storage periods with respect to the average one reduces possible influence of fading and ageing (see Section 2.2.8).

3.1.4. Material handling

To reduce ageing and fading, as well as the presence of scratches or dust on PADC, one should try to limit the PADC handling time before the monitoring period or between the end of the monitoring period and etching and follow the recommendations aforementioned for storage.

Another problem is the exposure to radon: even if the image software can easily discriminate the different types of tracks, it is important to limit it as much as possible. As radon exposure can be worsened by the generation of static electric charges on the detectors, one must avoid touching them: the use of gloves and/or anti-static spray is strongly recommended for the dosimeter assemble and disassemble stages, while a proper badge should avoid that the worker touch the dosimeter during its use. But most importantly, the badge should prevent radon from reaching the PADC detector at all (see also Section 3.2).

3.2. Badge design

The design of the badge is driven by several considerations. First, the badge design is limited by the geometry and space available for the neutron detector while keeping the badge convenient to wear. Because a photon field is almost always present, the whole dosimetry system must include a photon and a neutron detector.

In addition to these ergonomic considerations, there are also metrological considerations such as the energy range to be measured. If only the fast component ($\geq$ 100 keV) is considered, then a highly hydrogenated converter of a few mm will be sufficient. If the thermal component must be measured as well, then a second measurement area on the PADC with a thermal neutron converter must be added (see Section 2.3.1). The badge itself can be used as a converter, so that it is usually made of plastic materials (also because of weight and cost). If thermal neutron detection is required, particular plastic materials sensitive to thermal neutrons have to be selected (like polyamides, which contain ^{14}N).

There are also some constraints from the ISO 21909–1:2021 standard [48] that could drive the design of the dosimeter: the badge can be sealed for instance to prevent from radon exposure that can be a problem at very high levels. The ISO standard also gives tests and criteria for storage in harsh temperature and humidity conditions and one test and criteria to determine whether there is a change in the dosimeter response to neutrons due to physical damage (drop test).

Moreover, steps must be taken to guard against contamination. Contamination by α-emitters is a risk in some nuclear applications and a tiny amount of contamination can produce an appreciable false reading. Of course, as for radon, track shape/size discrimination may be possible, but it is better to prevent this scenario.

3.3. Quality assurance of the etching procedure

Because of the dependence of the final material sensitivity on the etching conditions, it is important to guarantee the quality and reproducibility of the etching.

The reagents should be controlled for dilution by titration or via hydrometers. The temperature of the etching solution should be controlled thermostatically and preferably recorded. Stirring is recommended throughout the entire etching process to guarantee thermal and chemical homogeneity.

Moreover, the entire etching step can be monitored by including among the detectors to be etched some reference irradiated detectors, which are then evaluated to check if the sensitivity was not affected by problems in the etching.

3.4. Dosimeter evaluation

3.4.1. Calibration and control detectors

For each monitoring period, calibration and background control detectors from the same PADC batch or sheet should be prepared and separated. These detectors should be etched and evaluated together with the routine detectors. It is important not to mix detectors from different batches not to risk applying the calibration factor obtained with one batch to dosimeters of a different batch.

Alternatively, one can sample and process the control detectors (calibration and background) before issuing the dosimeters and then apply software-controlled ageing/fading corrections when the dosimeters are returned, which deals with late returns effectively. That is why it is of importance to define the maximal limit for late-return dosimeter in its process and give such information to the customer.

The ISO 21909–1:2021 standard [48] defines specific test and associated requirements to determine whether fading and ageing are correctly accounted: for fading, the response should not be modified by $-15\%/+18\%$ between dosimeters analysed just after irradiation and some stored after irradiation for a period to be defined by the IMS. This period could be, for example, 120 days (equal to three months period +1 month for transit and analysis); a similar test and criteria are defined for ageing with a maximal period of storage before irradiation.

3.4.2. Investigation of doses

To discriminate false positives due to material defect and establish that a dose measured is real, further investigations can be performed. For example, sub-regions of the detector or the entire detector can be scanned. For a real neutron dose (assuming uniform neutron fluence on the PADC, as it happens in practical situations), the track density should be similar in all the sub-regions, whereas in case of material problems the track density will vary significantly across the detector surface. A statistical check (χ^2 test) can be performed to check if the track density is Poisson-distributed (see Section 2.3.3). If a fast neutron converter is used in front of the PADC, the PADC detector can also be scanned in the front and the back surface – for a real neutron exposure, the dose should not vary too much between these two measurements, as the PADC itself also works as a neutron converter. This assumption can easily be tested by applying the same procedure to calibration detectors.

For fast neutron fields, a thermal neutron converter placed above a region of the PADC surface can be useful in identifying whether a recorded dose is due to a genuine fast neutron exposure or to spurious high background; in fact, a true fast neutron exposure is always associated with a measurable thermal neutron dose at the body surface. Referring to this approach, the PADC region sensitive to thermal neutrons is sometimes denoted as a thermal neutron “tell-tale” [39].

False positives due to material problems tend to increase in time. For this reason, one should not use detectors that are too old; the exact period depends on the manufacturer and, therefore, must be specified by them. For the same reason, the monitoring period should also not be too long (<1 year).

3.5. Control of the entire process

3.5.1. Blind tests

Quality of the results can be monitored through blind tests, that is, including with each evaluation detectors from the same batch and handled in the same way as the routine detectors, but irradiated with a known dose. These tests may be nationally-required proficiency tests (e.g. in the UK, a "performance test", with a known source but unknown doses, must be undertaken at intervals of no more than 18 months), or in-house "dummy customer" tests.

3.5.2. Intercomparisons

IMSs participate routinely in neutron dosimetry intercomparisons. A national intercomparison is organized annually by the *Physikalisch-Technische Bundesanstalt* (PTB, Germany) and another one every three years by the *Institut de radioprotection et de sûreté nucléaire* (IRSN, France), whereas an international intercomparison is organized every five years by EURADOS. The results of the most recent EURADOS intercomparisons for neutron dosimeters, IC2012n and IC2017n, are reported in Refs. [27,56] and in a series of publications [20,28,55]. About 45% of the participants in the past EURADOS neutron dosimetry intercomparisons used PADC detectors.

4. Research needed

As clear from the discussion above, the successful use of PADC in neutron dosimetry requires a strict and time-consuming quality assurance program. Furthermore, there are several areas in which improvements are desirable:

(a) **Material consistency**: the consistent material production requires careful, sheet-by-sheet monitoring of the material properties, specifically background and sensitivity. It is not clear which quality assurance and quality control measures are important and are followed by the manufacturers.
(b) **Material sensitivity**: the material sensitivity can vary considerably from batch to batch and from manufacturer to manufacturer. It is important to identity which fundamental material parameters are correlated with the material sensitivity and how they vary as a function of manufacturing procedures. As discussed in (a), the goal here should be not only increase the sensitivity while keeping the background low, but also to improve the reproducibility of the sensitivity among different sheets and batches.
(c) **Intrinsic background**: The background impacts directly the detection limits. One must try to identify the main manufacturing parameters influencing the background to minimize it, without penalizing the sensitivity.
(d) **Ageing and fading**: as already mentioned, a major problem related to the PADC performances as a neutron dosimeter is that the material sensitivity varies in time and this effect is variable across sheets. One shall investigate how to reduce the impact of ageing and fading phenomena on the PADC material and also their variability. This can be done by acting "upstream", i.e. by properly modifying the PADC material composition looking for a material less susceptible to ageing and fading, and/or by acting "downstream", i.e. finding out novel and improved algorithms to successfully manage aged/faded detectors.
(e) **Improve the energy and angular response**: Although the PADC energy and angle response can be mitigated by the combination of dosimeter design and dosimetric techniques (using track-size-based correction factors, employing LET-based techniques, etc.; see Sections 2.4.1 and 2.4.2), it would be certainly of interest to study possible improvements in the PADC composition, in the etching procedures and in the signal analysis to investigate if a dosimeter with an enhanced response is achievable.

Therefore, research is needed along the following lines:

(a) investigating and comparing the currently available PADC materials;
(b) relating the physical and dosimetric properties of a given material to its composition and manufacturing process;
(c) seeking for improvements in the production process and quality assessment tests to reach an optimized PADC for neutron personal dosimetry applications.

A close collaboration with the manufacturers is essential because of their expertise and the fact that manufacturing details are kept confidential for commercial reasons.

Because the final neutron dose estimate depends on a combination of parameters, not all of which can be separated or controlled, the comparison of different materials is not trivial. The interplay among the different components of the dosimetry system also makes it difficult to isolate the contribution of specific components to the final dosimetric properties of the system. One shall try to investigate which PADC material properties can be defined and characterized independently of the dosimetry system, to be used when comparing the quality of different materials and how these properties can be translated into dosimetric performances. More specifically, the final neutron dose estimate depends on the combination of several parameters which can be grouped in four categories: i) intrinsic quality of the PADC; ii) dosimeter structure, in particular the presence of a converter; iii) etching procedure; and iv) track reader and track discrimination algorithms. As the scope of this investigation is the quality of the PADC, i.e. point (i), it is quite challenging to keep under control the parameters (ii) - (iv). The only feasible approach is to define a reference badge, a reference etching condition and a reference reader, possibly with a full control on the algorithm of analysis, adjustable for the specific material.

5. Final remarks

PADC remains one of the main types of detectors for neutron dosimetry, with satisfactory results demonstrated in international intercomparisons. Nevertheless, research is needed to improve the PADC material consistency and sensitivity, while keeping the background low or reducing it, as well as to minimize the effects of ageing and fading and improve the energy and angular response. This requires considerable efforts to investigate and compare available PADC materials, relate the dosimetric properties with the composition and manufacturing parameters, and finally improve the production processes. This can only be achieved by a closer collaboration between researchers, IMS and PADC manufacturers. Due to the large effort involved, it is unlikely that major advances can be achieved isolated, justifying the coordinated effort promoted by the EURADOS Working Group 2, of which this work is the first step. Thus, with this review we hope not only to help improve the PADC-based neutron dosimetry by sharing best practices, but also to establish a common framework for the next steps.

CRediT authorship contribution statement

M. Bolzonella: Conceptualization, Writing – original draft, Writing – review & editing, Visualization. **I. Ambrožová:** Conceptualization, Writing – original draft, Writing – review & editing. **M. Caresana:** Conceptualization, Writing – original draft, Writing – review & editing, Supervision, Funding acquisition. **N. Gibbens:** Conceptualization. **P. Gilvin:** Conceptualization, Writing – original draft, Writing – review & editing, Supervision. **F. Mariotti:** Conceptualization, Writing – original draft, Writing – review & editing. **A. Savary:** Conceptualization. **A. Stabilini:** Conceptualization, Writing – original draft, Writing – review & editing, Visualization. **F.A. Vittoria:** Conceptualization, Writing – original draft, Writing – review & editing. **E.G. Yukihara:** Conceptualization, Writing – original draft, Writing – review & editing,

Visualization, Supervision, Funding acquisition. **M.-A. Chevallier:** Conceptualization, Writing – original draft, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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