ELECTROCHEMICAL ETCHING PROCESSES FOR THE DETECTION OF NEUTRONS AND RADON-DECAY PRODUCTS

by

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The electrochemical etching, because of its complexity, is of interest when it makes it possible to achieve detection characteristics which are not encountered with the chemical etching. These unique characteristics can be found for example for the personal dosimetry of low-energy neutrons around nuclear reactors and for the detection of both low- and high-energy cosmic-ray neutrons at civil aviation altitudes. In particular, sufficiently large signal-to-noise ratios for cosmic ray neutron measurements can be achieved by using stack of polycarbonate- and/or CR-39-detectors, since the electrochemical etching processes make it possible: (a) the rapid scanning of large detector areas, and (b) the counting of coincidence events in paired detectors induced by a-few-microns long tracks.

The detection of the radon decay products is hindered by the fact that their concentrations are altered in the vicinity of detector surface during the measurement. Polycarbonate detectors may be useful in solving these problems both because they register radon-decay products far away from the plated-out surface and they can be manufactured with any possible geometry and/or shape. However, it is possible to use several combinations of chemical and electrochemical etching steps which implies the possibility of new applications of track detectors for the registration of neutrons, cosmic rays and radon decay products.

Key words: radon, neutrons, cosmic rays, track detectors, dosimetry, chemical etching, electrochemical etching

INTRODUCTION

The most important incentive for the development of electrochemical etching (also referred to as ECE) was the urgent need to improve neutron dosimetry [1]. The personal fast neutron dosimeter available in the 1970's was based on nuclear emulsions [2], which relied on detection of the tracks of individual recoil protons in thin photographic emulsions. This dosimeter manifested various shortcomings such as poor low-energy response, track fading and sensitivity to low-energy gamma- and X-rays. These drawbacks were very severe in the case of personal neutron dosimetry in facilities having large numbers of low-energy neutrons and mixed gamma neutron fields, such as reactors [2].

Soon after the discovery of the electrochemical etching, it was possible to develop a personal neutron dosimeter based on neutron-induced recoils in polycarbonate (PC) detectors, whose response was unaffected by X-ray or gamma radiation, by fading or by environmental parameters, such as high temperature and humidity [2, 3]. This detector was quite promising, so that, soon after its discovery, a commercial dosimetry service adopted it for the use in routine personnel dosimetry [2]. However, with this dosimeter it was possible only to register doses of neutrons with energy larger than 1 MeV. It was the discovery of CR-39 [4] that made it possible to detect neutron-induced proton recoils with energy as low as tens of keV [5, 6].

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DOSIMETRY OF LOW-ENERGY NEUTRONS

To understand the importance of electrochemically etched detectors for neutron dosimetry it is useful to describe the differences between the chemical-and electrochemical etching processes.

For example, fig. 1 illustrates the differences in response between these two etching processes for the particular case low-energy proton recoil tracks, assuming that they originate from the surface.

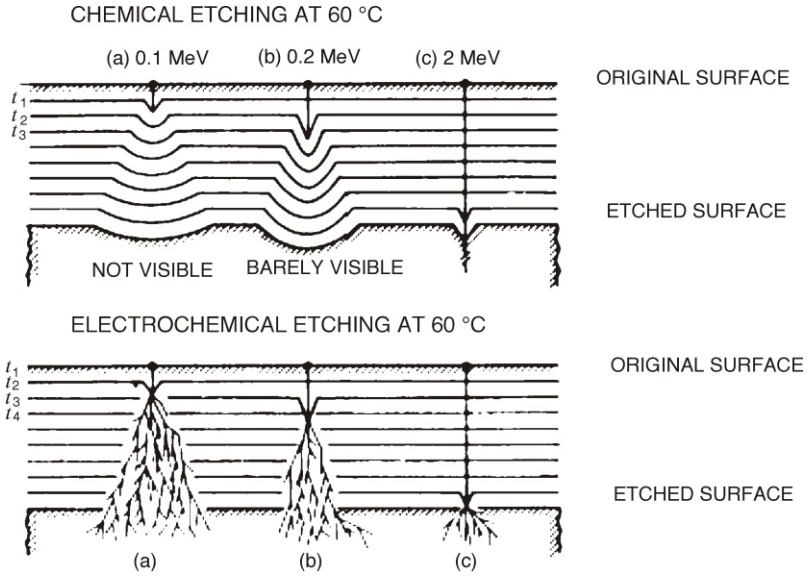

Figure 1. Schematic illustration of chemical and electrochemical etching processes

The track formation is shown schematically at different etching times t_1, t_2, t_3, *etc*., [5]. In the case of chemical etching (upper part of the figure), tracks (a) and (b) remain pointed until they are etched to the ends of their trajectories. Since prolonged etching is usually required in order to be able to etch high-energy protons, track (c), tracks (a) and (b) both become blurred and may disappear from view.

At the end of etching, thick detector layers (typically 5-15 μm thick) have been etched away from the original surface and most of the short-range tracks within these layers may have disappeared from view or are very difficult to count. This is particularly relevant for neutron energies below 500 keV, where, for a single dosimeter, large variations in the number of counted tracks occur among different observers and, for a given observer, at different times of counting [5, 6].

The lower part of fig. 1 shows the initiation and propagation of treeing phenomena in newly formed tracks for purely electrochemical etching processes. In this case, once a track with a pointed shape has been formed, it starts treeing. As the electrochemical etching proceeds, those tracks already treeing, (a) and (b), continue to enlarge while more tracks are formed and initiate treeing, track (c). This type of etching can be considered as just the opposite of the chemical etching at the same temperature, since low-energy recoil tracks keep enlarging and becoming more visible under electrochemical etching processes, while they may disappear from view under conventional etching as shown in the upper part of fig.1. If tracks are formed at the beginning of electrochemical etching processes, they will grow to a greater extent than those which will be formed at a later time, thus resulting in very different spot diameters [7, 8]. The response to protons of CR-39 detectors was thoroughly investigated by Cross *et al*. [6], who demonstrated that the electrochemical etching procedure can simultaneously detect protons from 10 keV to 3 MeV. Figure 2 shows track spots produced by protons with energies of 30, 50, 100, and 145 keV respectively [5]. On the basis of these results Cross *et al.,* [6] calculated the response of CR-39 to neutrons. The results of this calculation are shown in fig. 3 [6]. The solid line shows the number of tracks per cm^2 per 10 μSv calculated for CR-39 with a 2 mm thick polyethylene radiator in front of it. The dashed curves show the separate contributions of protons generated in the etched layer, which predominate at low energies, and protons from the radiator which predominates above 1 MeV. The points were measured for normally-incident monoenergetic neutrons. All the above results were obtained by using a 25 $kVcm^{-1}$ rms electric field at 60 Hz with 6 N KOH in water for 5 h at 60 °C [6].

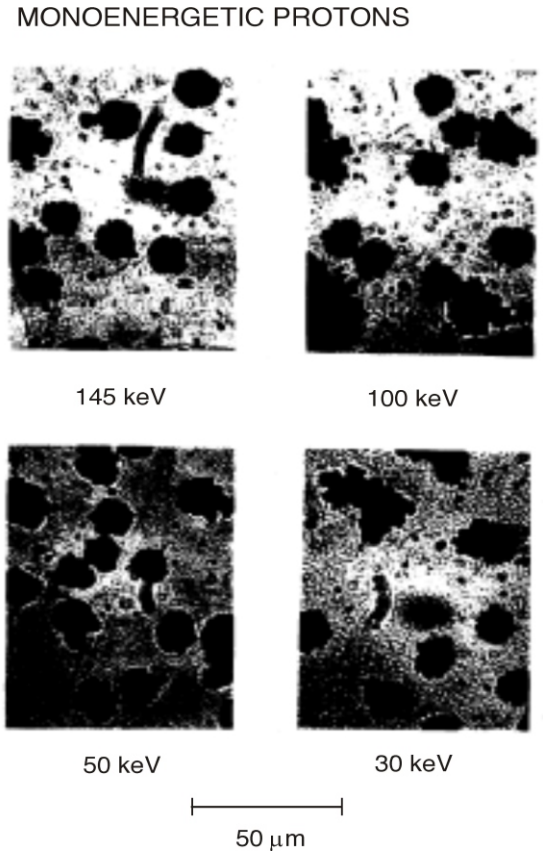

Figure 2. Electrochemically etched proton-induced tracks

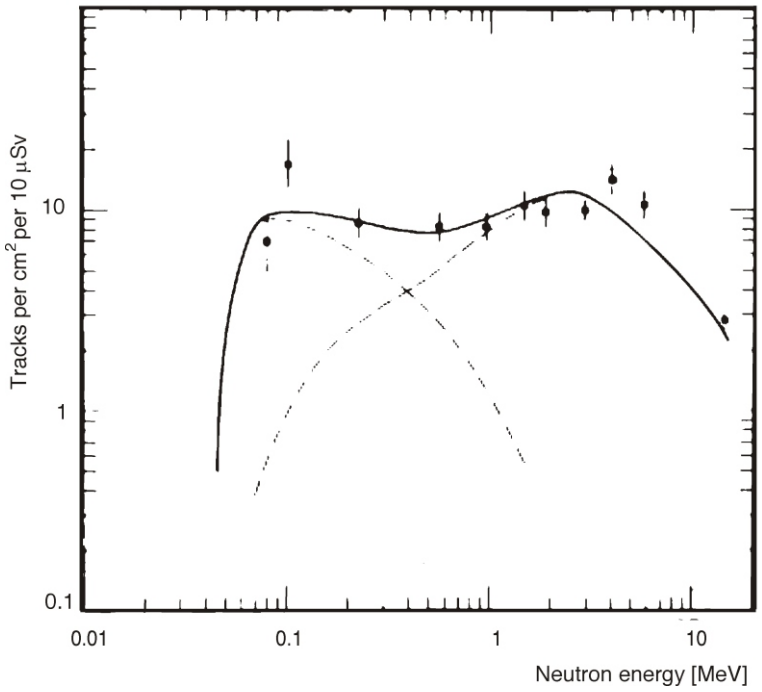

Figure 3. The response of electrochemically etched CR-39 detectors versus neutron energy

DOSIMETRY OF COSMIC-RAY HIGH-ENERGY NEUTRONS BY ECE DETECTORS

With the complex fields present at flight altitudes and/or around high energy accelerators, the registration of sufficiently low neutron doses by damage track detectors is hampered by the poor signal-to-noise ratio of these detectors.

The simplest way to increase the registration sensitivity of damage track detectors is to increase both the exposure time and the detector area. Because of their unique storage property, damage track detectors can be exposed for any desired length of

time. On the other hand, the registration of large areas (hundreds of cm^2) can easily be carried out by electrochemical etching detectors. However, the possibility to increase the registration sensitivity by increasing the detector area is limited by the strong variability of the background of track detectors.

The background problems were finally solved by a new registration method based on counting coincidence spots in a matched pair of detectors [9]. By using electrochemical etching, coincidence spots can easily be induced even by a few-microns-long track in paired detector-surfaces.

The coincident method is well established for long-range particles which cross different track detectors in a stack (upper part of fig. 4). This coincidence method has been successfully exploited for the registration of high charge and energy (HZE) particles in cosmic rays since the early discovery of damage track detectors [10].

A completely new coincidence method is proposed in the lower part of fig. 4, where the etched-track ranges are so short that they do not cross any of the detectors, but they just penetrate both of the matched detector surfaces. These tracks are typically produced by neutron-induced recoils in PC and CR-39 detectors [9]. It is obvious that the only way to produce coincident spots with a few-micron long tracks (which can be easily counted in large areas) is through the electrochemical etching processes.

The upper part of fig. 5 shows coincidence events produced by electrochemical etching spots on two paired polycarbonate detectors as they appear in a microfiche reader. The identification of coincidence events is very simple, in spite of the poor image. The lower part of the figure shows a pair of electrochemical etching spots, which do not form a true coincidence since they belong to the same detector surface: they have the same contrast and present the typical interacting characteristics of close electrochemical etching spots.

When compared with the responses of detectors based on counting tracks on a single surface, the response of the detectors based on the coincidence-method present the following advantages [9]:

(1) consistently low background,
(2) relatively flat response, and
(3) detection with different neutron-energy thresholds.

COMBINATION OF CHEMICAL- AND ELECTROCHEMICAL ETCHING PROCESSES FOR NEUTRON AND RADON DOSIMETRY

The most attractive characteristics of electrochemical etching processes involve the possibility of using a variety of combinations of etching parameters and/or chemical and electrochemical etching processes for the purpose of enhancing the desired detector response. This can easily be achieved by controlling the etching procedure with an external apparatus. Bartlett and Steel [11] suggested the use of a microprocessor-controlled combination of chemical- and electrochemical- etching processes to provide a reasonably low background and an acceptable energy response for neutron dosimetry.

In practice, by proper choice and control of the electrochemical etching parameters in CR-39 during etching, it is possible to expand considerably the usefulness of the electrochemical etching pro-

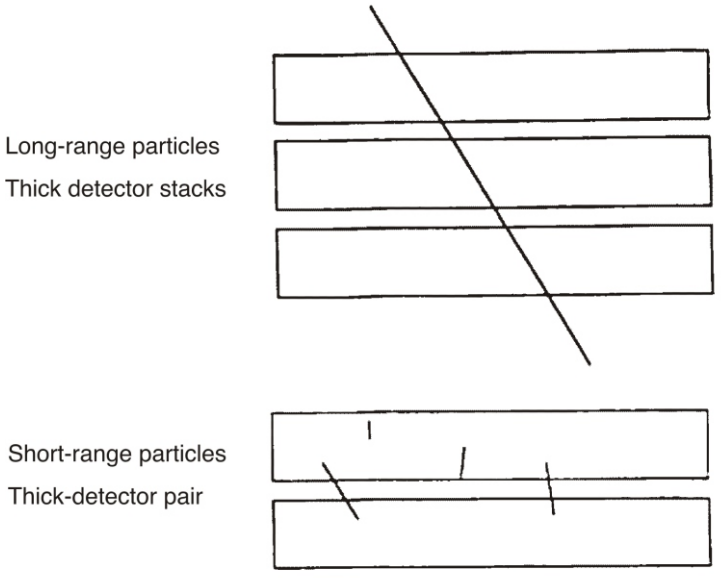

**Figure 4. Upper part: coincidences induced by long-range particles.
Lower part: coincidences induced by short-range particles**

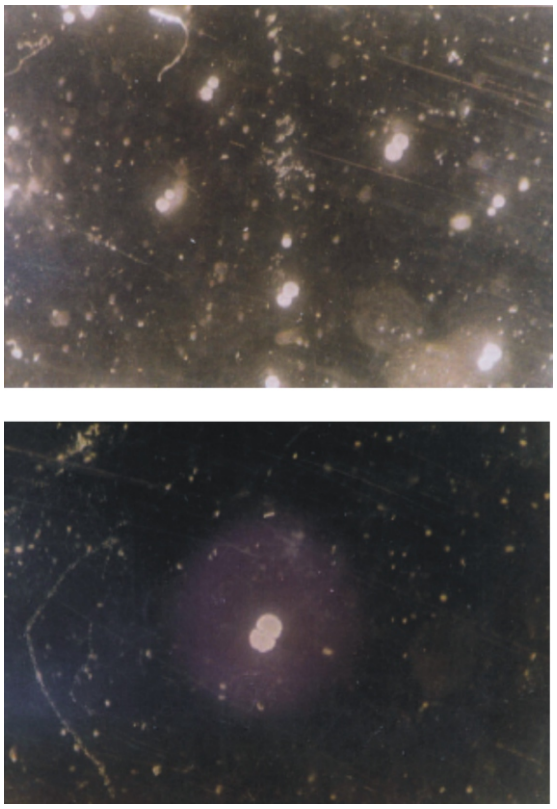

Figure 5. Paired spots induced by neutron recoils in electrochemically etched polycarbonate

cesses [12] both for the detection of neutrons, radon, and its decay products.

Track formation by chemical etching prior to electrochemical etching

The electrochemically processed detectors can successfully be used when track density is less than a few thousand tracks per square centimetre. For higher track densities both the spot diameter and electrochemical etching efficiency decrease as the track density increases [13]. However, a chemical etching step for track formation prior to the electrochemical etching may be useful to increase the response linearity [14]. Figure 6 shows electrochemically etched tracks of alpha particles from radon on CR-39 [15], where the track formation has been obtained by a chemical pre-etching. When the CR-39 foil is observed under an optical transmission microscope, tracks appear as dark spots, as shown on the left-hand side of fig. 6. By focusing close to the surface (especially with a reflection type of microscope) the electrochemically etched spots will disappear and only chemically etched tracks appear, as clearly illustrated on the right-hand side of fig. 6. The possibility to separate the chemically-etched tracks from the electrochemically etched tree-spots makes it feasible to exploit the advantages of both types of etching.

Electrochemical etching followed by chemical etching

When using electrochemically etching track detectors for neutron dosimetry in space, it is important to differentiate the response to neutrons (and protons) from that of heavy charged particles (HZE particles). This can be simply carried out by a two steps etching. A first step of electrochemical etching is carried out only on one detector surface for rapid scanning of large areas. To identify the spots due to HZE particles, Bartlett *et al*., [16] first proposed to use a second step consisting of chemical etching only. Because of the large range of the HZE, an etched pit is produced on the non electro-

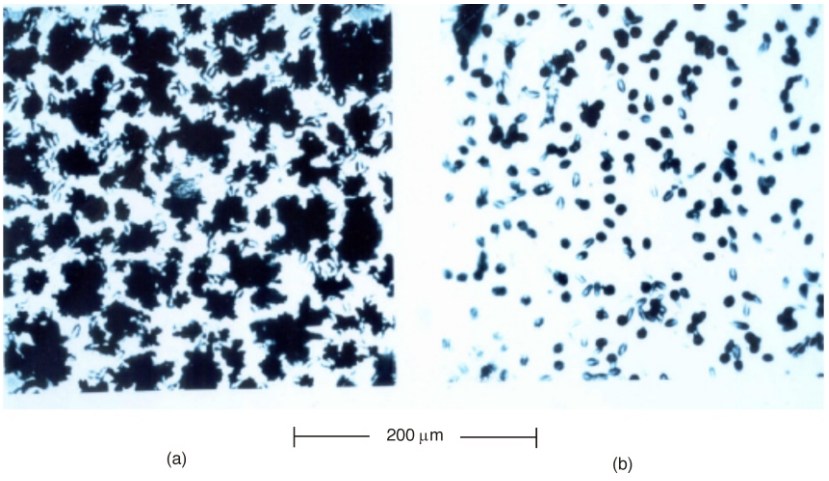

Figure 6. Chemically- plus electrochemically etched tracks: (a) treeing components, (b) chemical-track components. For viewing conditions, see text

chemically etched surface and also at the back of the electrochemically etched pit, allowing track of HZE to be distinguished from tracks of neutron secondaries [16].

CR-39 WITH NO RESPONSE TO THE PLATED-OUT RADON-DECAY PRODUCTS

Because of its high sensitivity, CR-39 registers full-energy alpha particles from radon decay products plated-out on its surface, while the cellulose nitrate and the polycarbonate detectors are both insensitive to full-energy 's from plate-out. These latter characteristics are very important for the measurement of radon decay products or, in general, when using track detectors in the bare configuration [17]. However, CR-39 could be made insensitive to plate-out, if processed in such a way that the tracks of alpha particles (with energies of 6 and 7.7 MeV) from radon daughters are not revealed. This can be achieved by a two-steps etching consisting respectively of a chemical etching step for the formation of only those tracks with energy below 6 MeV, followed by an electrochemical etching step to enlarge only those tracks which have been formed [14, 18].

Table 1 shows the registration efficiency of different bare detectors (LR-115, PC, and CR-39) exposed at the NRPB radon chamber to 356 kBqm^{-3}h of radon gas at an equilibrium factor of F = 0.4. The etching conditions are:

– Chemical etching for 90 minutes in 10% (by weight) of NaOH in water at 60 °C for LR115;

– 5 hours chemical pre-etching with PEW solution at 60 °C plus electrochemical etching at 30 °C in 30% KOH in water for 5 hours at 30 [kVcm^{-1}] rms and 2 kHz for polycarbonate. In the case of CR-39, the electrochemical etching processes are the same of those for polycarbonate, while different types of chemical pre-etching were used, such as the following:

– 5 hours chemical pre-etching at 24 °C with 30% KOH in water,

– 10 hours chemical pre-etching at 24 °C with 30% KOH in water, and

Table 1. Registration sensitivity of different detectors

Detector type	Sensitivity [cm^{-2} kBq^{-1} h^{-1}]
LR–115	1.10 0.13
Polycarbonate	0.52 0.05
CR–39 (5 hours pre-etching)	0.80 0.11
CR–39 (10 hours pre-etching)	3.0 0.4
CR–39 (heavy pre-etching at 70 °C)	6.6 1.6

– 5 hours chemical pre-etching at 70 °C with 30% KOH in water.

While the 5-hours chemical pre-etching at 70 °C induces the formation of all alpha-particle tracks from radon-decay products plated-out on the CR-39 surface, the registration of these alpha particles does not occur with the other types of pre-etching conditions, listed above for CR-39.

These latter etching conditions make it possible to register only particles with energy less than 5 MeV [14, 18] and thus alpha particles from plated-out daughters are not registered. In contrast with the shortcomings of CR-39 etched only chemically (being sensitive to plate-out), the CR-39 may prove to be the most interesting detector when both chemical and electrochemical etching processes are concerned. In this case, it is possible to obtain a detector with all the characteristics required (including the same of those of LR-115 and PC) by an appropriate choice of the chemical etching conditions prior to the electrochemical etching.

The polycarbonate detector has the lowest registration sensitivity. In fact, this is the property which makes this detector very valuable for the detection of radon decay products, as described in the following.

THE UNIQUE CHARACTERISTICS OF ELECTROCHEMICALLY ETCHED POLYCARBONATE DETECTORS

It is well known that the plate-out of radon decay products on surfaces of different materials causes a depletion of their concentration in the surroundings of all the material-surfaces, which depends on many factors such as surface-area, environmental parameters, electrostatic charges, ventilation rate *etc*., [19-22]. This depletion occurs also in the vicinity of the detector surface, thus affecting what is intended to be measured. For a given atmosphere of radon and radon decay products, this effect increases with the area of the detector surface. Eventually the depletion of radon decay products decreases as the distance from the detector-surface increases [23-24]. Bigazzi *et al*., [20] and Hadler *et al*., [21] claim that the depletion occurs mostly within 3 cm from the detector surface.

Since electrochemically etched polycarbonate foils register only alpha particles with energy less than about 2 MeV, they could detect only radon decay products at a distance greater than about 4 cm from their surfaces. The ideal detector for this application would be a sphere made of polycarbonate which could easily be manufactured or are already available as a consumer-type of product. This sphere could also be an excellent detector for neutrons, since the major problem with the damage track detectors for neutron dosimetry is the strong angular depend-

ence of their response. Incidentally, the problem of the strong angular dependence of the polycarbonate neutron dosimeter could be solved by using [25] electrochemically etched polycarbonate test tubes or small bottles. These container types of detectors could be of interest also for their use as bare detectors for radon and its decay products. It is possible to have detectors of any shape (often already available as consumer product), which makes the electrochemical etching of polycarbonate a detector with unique characteristics.

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Луиђи ТОМАСИНО

ПРОЦЕСИ ЕЛЕКТРОХЕМИЈСКОГ РАЗВИЈАЊА ДЕТЕКТОРА ЗА РЕГИСТРОВАЊЕ НЕУТРОНА И ПРОДУКАТА РАДИОАКТИВНОГ РАСПАДА РАДОНА

Услед своје сложености, електрохемијско развијање од интереса је када омогућава да се достигну својства детекције која хемијско развијање не обезбеђује. Ове јединствене околности могу се, на пример, сусрести у личној дозиметрији нискоенергетских неутрона у околини нуклеарних реактора, али и при детекцији како нискоенергетског тако и високоенергетског космичког зрачења неутрона на висинама које одговарају цивилном авионском саобраћају. Посебно, довољно велики однос сигнала и шума при мерењу космичког неутронског зрачења може бити остварен коришћењем поликарбонатног пакета или CR-39 детектора, будући да процеси електрохемијског развијања омогућавају да се изврши: (а) брзо скенирање пространих детекторских површина и (б) пребројавање помоћу детекторских парова коинцидентних догађаја изазваних траговима од тек неколико микрона.

Детекција продуката радиоактивног распада радона отежана је услед промене њихове концентрације током мерења у околини површине детектора. Поликарбонатни детектори могу бити корисни у решавању ове тешкоће тиме што бележе продукте радиоактивног распада радона даље од спољне површине детектора и што се могу произвести у различитој геометрији и облику. Шта више, могуће је користити већи број комбинација хемијских и електрохемијских корака у њиховом развијању што наговештава нове примене траг детектора за регистровање неутрона, космичког зрачења и продуката радиоактивног распада радона.