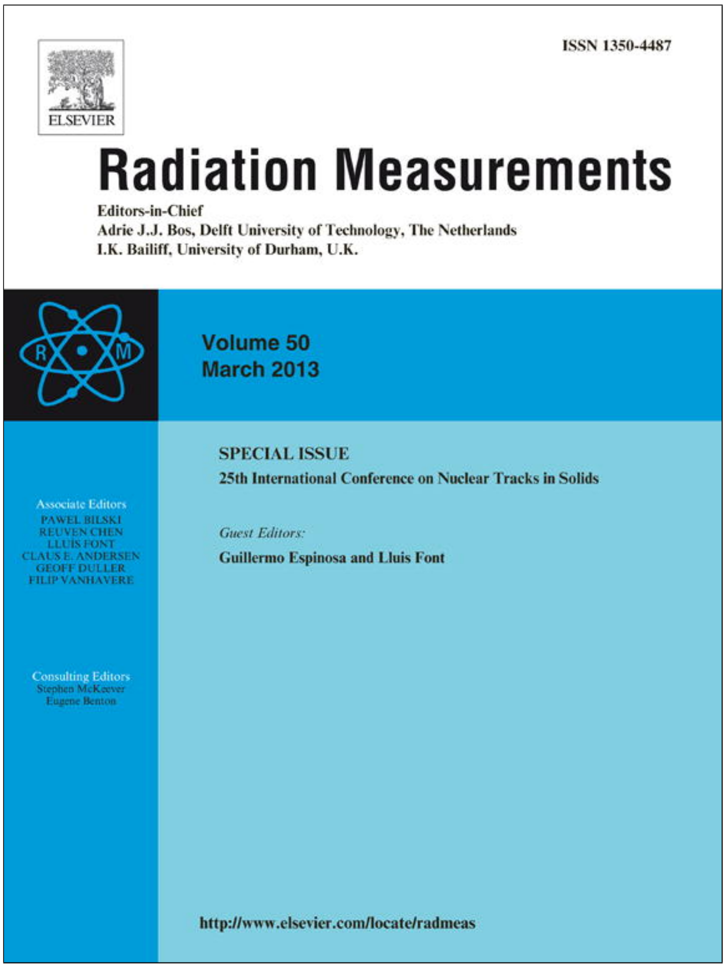

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# Experimental study on the effect of high humidity environments on the response of long-term exposed nuclear track detectors

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## HIGHLIGHTS

► We expose the nuclear track detectors Makrofol at different high humidity environments.
► Humidity effect over the nuclear track detectors Makrofol depends on exposure time.
► We compare different configurations of polyethylene filters and bags to protect the detector.
► We analyse the influence of the polyethylene membrane on detector response.

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## ABSTRACT

In Spain, a recent modification in the regulations of protection against ionizing radiation obligates to determine radon levels in particular workplaces like spas, mines and caves. Most of these workplaces may present extreme measurement conditions like high aerosol content, temperature, and humidity that can affect the radon detector response. In our laboratory we use to determine mean radon activity concentration in air with the nuclear track detector Makrofol DE covered with an aluminized Mylar foil and placed within the FzK FN diffusion chamber with a glass fibre filter. We have compared detector response using different filter configurations in common laboratory ambient conditions. The configuration with a better response (polyethylene filter) and our reference configuration took part in a study of the effect of different temperature and humidity on our detector response carried out in the INTE radon chamber. Results obtained did not show a significant difference between detector responses with both filters. However, when we exposed them for long periods to real environmental conditions at underground sites we could observe Mylar deterioration. To look in detail into a possible effect of long-term high-humidity exposures we exposed nine sets of detectors with three different polyethylene bags, first five sets under controlled conditions in the INTE radon chamber and then four sets in long-term exposures at high humidity environments. We have seen that the Mylar foil can be damaged depending on the duration of exposure. In a radon chamber exposure time is normally limited to a few days for practical and financial reasons; therefore, results do not show if humidity affects the glass fibre filter and detectors response. To analyse it we exposed detectors in a real humid environment up to a month where we already observed Mylar deterioration due to humidity but the possible impact on track density is hidden by the scattering of the results found, so a clear conclusion cannot be stated.



## 1. Introduction

In 2001, natural radiation was considered for the first time in Spanish regulations of protection against ionizing radiation (RD 783/2001, 2001). With the recent modification of these regulations (RD 1439/2010, 2010) owners of workplaces with natural radiation sources are required to declare their activities and to carry out studies to check if workers have an increment of exposure that could be significant from a radiological protection point of view. Among workplaces that must be studied there are those where workers can be exposed to a significant exposure due to thoron and radon daughters like spas, mines, caves, underground workplaces and workplaces at identified areas. At some of these workplaces there are environmental conditions that could affect detector response, like high aerosol concentration, high temperature and high humidity.

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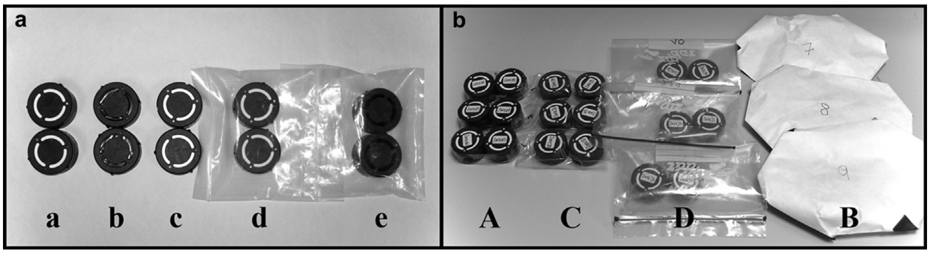


**Fig. 1.** (a) Configurations of filters and polyethylene bags analysed in the first phase of the study. (b) Configurations of polyethylene bags analysed in the second phase of the study.

In the last years we have measured radon levels in several workplaces (Font et al., 2008) where measurement conditions were already extreme so we could observe how our standard detector (with a glass fibre filter) was affected after long-term exposures. For instance, in some places the filter was covered with a lot of dust or in very humid environments, the Mylar layer and the glass fibre filter presented deterioration.

Polyethylene membranes have been widely used to prevent from high humidity. For instance, Azimi-Garakani et al. (1988) developed a passive radon detector that was heat-sealed in a bag of 40 μm thick polyethylene; the Health Protection Agency (HPA) of Chilton, UK, adopted the method of sealing standard area radon detectors in 200 μm thick polyethylene for measurements in areas with high humidity (Miles et al., 2009), and Tommasino et al. (2009) solved the problem of humidity enclosing their radon badge in polyethylene bags characterized by a large permeability to radon and a small permeability to water vapour. We decided to carry out a specific study for optimizing our track etched detector response in high humidity environments using polyethylene membranes.

## 2. Methodology

The detector we use to determine radon concentration is the Karlsruhe FN dosimeter based on Makrofol DE (Baixeras et al., 1996). Makrofol is a polycarbonate very widely used as nuclear track detector. Makrofol foils of 500 μm thick are covered with an aluminized Mylar foil of 3 μm thick, which avoids the creation of static charge on their surfaces that may enhance the deposition of radon decay products. A glass fibre filter prevents dust and radon progeny to enter into the chamber. After exposure, the Mylar foil is removed, and the Makrofol foils are electrochemically etched and analyzed in our laboratory (Amgarou et al., 2003). This detector was calibrated in the HPA radon chamber and we participated in different national and international comparisons (Moreno et al., 2008). The problem with humidity mentioned above has been studied in two separated phases with different configurations of polyethylene membranes. In the first phase we prepared five configurations with polyethylene films used as internal filter or as external bag (see Fig. 1(a) and Table 1). In the second phase of the study three polyethylene bags of different commercial brands (see Fig. 1(b) and Table 1) have been analyzed. In both phases, the first configuration (a and A) corresponds to our reference configuration. At each phase we have exposed these configurations at different environmental conditions. In the first phase at the common temperature and humidity conditions of our laboratory, then at controlled conditions inside a reference radon chamber, and finally at real environmental conditions inside different underground indoors. In the second phase we exposed them to the radon chamber and then in an underground workplace.

**Table 1**
Characteristics of the different configuration of filter and bag studied in separated phases.

| Phase | Conf. code | Filter | Thickness (mm) | Bag | Thickness (mm) |
|---|---|---|---|---|---|
| I | a | Glass fibre | 350 ± 20 | – | – |
| | b | Polyethylene | 37 ± 2 | – | – |
| | c | Glass fibre + Polyethylene | 387 ± 20 | – | – |
| | d | Glass fibre | 350 ± 20 | Polyethylene | 62 ± 2 |
| | e | – | – | Polyethylene | 62 ± 2 |
| II | A | Glass fibre | 350 ± 20 | – | – |
| | B | Glass fibre | 350 ± 20 | Tyvek | 115 ± 6 |
| | C | Glass fibre | 350 ± 20 | Treseses | 33 ± 2 |
| | D | Glass fibre | 350 ± 20 | Zipdar | 51 ± 2 |

### 2.1. Laboratory conditions

In our laboratory we have a small radon box of PVC that we use for research and internal quality control (Amgarou, 2002). We can reach radon concentrations of the order of kBq $m^{-3}$ by means of small samples of pitchblende. We monitor radon continuously with an active detector like PRASSI (Silena[1]), Rad 7 (Durridge[2]), AlphaGUARD (Saphymo[3]), or ATMOS 12 (Landauer Nordic AB[4]). Inside this box we have exposed for 5 days the first 5 configurations (8 detectors of each configuration).

### 2.2. Controlled conditions

The exposures have been done at the Institute of Energy Technologies (INTE) of the Polytechnic University of Catalonia at Barcelona, Spain, where there is a homologated radon chamber (Vargas et al., 2004). This chamber has an automatic control of environmental parameters and radon concentrations and it is used for calibrations and intercomparisons. All characteristics of exposure carried out in this chamber (radon concentration, exposure time, temperature, humidity, number of configurations analyzed and total number of detectors exposed) are presented in Table 2. Exposures of the first phase correspond to an intercomparison exercise we have taken advantage for in our study. But due to space constrains, we could expose configurations a and b only (Vargas and Ortega, 2007). Exposures of the second phase have been carried out exclusively for the purpose of this study (Moreno et al., 2011). The duration of all exposures has been limited to a few days for practical and financial reasons.

[1] Silena S.p.A, Via Firenze, 3, 20063 Cernusco s/n, Milano, Italy.
[2] DURRIDGE Company, Inc., 524 Boston Road, Billerica, MA 01821, EEUU. www.durridge.com.
[3] Saphymo GmbH, Heerstrabe 149, D-60488 Frankfurt a.M., Alemanya. www.genitron.de.
[4] Landauer Nordic AB, Vallongatan 1, SE-752 28 Uppsala, Suècia. www.gammadatainstrument.se.

**Table 2**
Characteristics of all exposures carried out inside the INTE radon chamber in separated phases.

| Phase | Expo code | $C_{Rn} \pm 2\sigma$ (kBq m$^{-3}$) | $T$ (°C) | Hr (%) | Time (h) | Number of configurations analyzed | Total number of detectors |
|---|---|---|---|---|---|---|---|
| I | 1 | 8.3 ± 1.2 | 20 | 45 | 76 | 2 | 6 |
| | 2 | 8.5 ± 1.2 | 20 | 30 | 72 | 2 | 6 |
| | 3 | 8.3 ± 1.2 | 10 | 45 | 66 | 2 | 6 |
| | 4 | 9.5 ± 1.3 | 30 | 45 | 71 | 2 | 6 |
| | 5 | 8.9 ± 1.3 | 20 | 80 | 71 | 2 | 6 |
| | 6 | 9.8 ± 1.4 | 30 | 80 | 69 | 2 | 6 |
| II | 7 | 8.5 ± 1.2 | 20 | 45 | 74 | 4 | 24 |
| | 8 | 17.0 ± 2.4 | 20 | 45 | 67 | 4 | 24 |
| | 9 | 20.0 ± 2.8 | 20 | 45 | 50 | 4 | 24 |
| | 10 | 20.0 ± 2.8 | 20 | 85 | 50 | 4 | 24 |
| | 11 | 20.0 ± 2.8 | 30 | 90 | 50 | 4 | 24 |

**Table 3**
Characteristics of all exposures carried out inside the ancient underground mines in separated phases and the radon levels obtained monitoring continuously radon concentrations with the AlphaGUARD monitor at some mine levels (level −3 at exposure 2 and level −2 at exposure 4).

| Phase | Expo | Season | T. (°C) | Hr. (%) | Mine level | Time (day) | Num. det. | $C_{Rn} \pm 2\sigma$ (kBq m$^{-3}$) |
|---|---|---|---|---|---|---|---|---|
| I | 1 | Summer | 17 | 80–100 | −1, −2, −3 | 7 | 26 | – |
| II | 2 | Spring | 14 | 80–100 | 0, −1, −2, −3 | 4 | 17 | 8.4 ± 0.3 |
| | 3 | Summer | 20 | 89 | −2 | 19 | 20 | – |
| | 4 | Summer | 20 | 89 | −2 | 10 | 20 | 10.8 ± 0.3 |
| | 5 | Summer | – | – | 0 | 29 | 20 | – |

### 2.3. Real conditions

In the first phase of the study we have exposed our reference configuration (a) and the configuration b inside two different underground places: (i) a private basement excavated into soil and (ii) underground ancient mines. In the second phase we exposed configurations A, B, C and D in the environmental conditions of the underground ancient mines only.

The private basement is located at the volcanic region of La Garrotxa, close to one of the blowholes analysed in another specific study (Moreno et al., 2009). Inside this basement radon concentration and relative humidity present very important seasonal variations, from 3 kBq m$^{-3}$ and 40% in summer to 0.2 kBq m$^{-3}$ and more than 90% in winter, respectively. We measured radon concentration for three years with exposure times from five weeks to four months.

Nowadays the ancient underground mines are both an archaeological site and a museum. Inside these mines humidity is

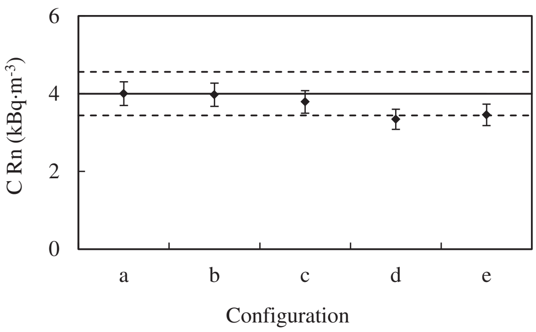


**Fig. 2.** Radon concentration obtained with the configurations a, b, c, d and e (see Table 1) exposed inside a radon box at habitual environmental conditions. Error bars correspond to standard deviation of the mean of 8 detectors, solid line is the reference value and dashed lines correspond to the standard deviation of the mean ($k = 2$).

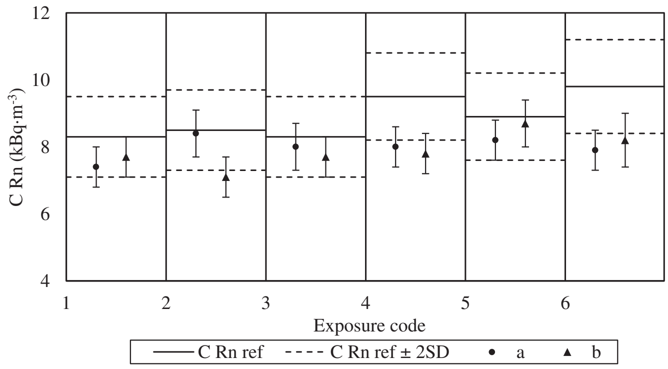


**Fig. 3.** Radon concentration obtained with the configurations a and b (see Table 1) exposed inside the INTE radon chamber at different controlled environmental conditions (see Table 2). Error bars correspond to standard deviation of the mean of 3 detectors, solid line is the reference value and dashed lines correspond to the standard deviation of the mean ($k = 2$).

kept high for the whole year, over 80%. We measured radon concentration at each station for a week (Font et al., 2008) finding out that radon levels present very important seasonal variations with the highest values in summer as it has been observed in other underground sites (Moreno et al., 2009, 2008; Kávási et al., 2006). These mines are an excellent natural laboratory to check our radon detectors for this study because there are different underground levels with different radon concentrations. In addition, we have the permission from the authorities to measure for long periods of time. Exposure conditions in these mines in both phases of the study are presented in Table 3. Environmental conditions and radon concentration have been monitored continuously by means of the AlphaGUARD monitor, which has an internal weather station and it is not affected by humidity.

## 3. Results and discussion

### 3.1. First phase

In phase I detectors were exposed inside our radon box at common laboratory conditions. In Fig. 2 it may be seen that all configurations present similar response, taking into account the uncertainty bars. Configuration b shows a mean value closer to our reference configuration (a) response than all other configurations; therefore, configurations a and b have been selected to be exposed

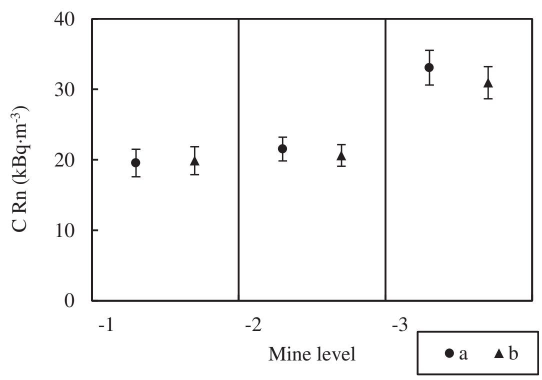


**Fig. 4.** Radon concentration obtained with the configurations a and b (see Table 1) exposed inside underground mines at high humidity conditions and different mine levels. Error bars correspond to standard deviation in level −1 and standard deviation of the mean of three detectors in level −2 and of two detectors in level −3.

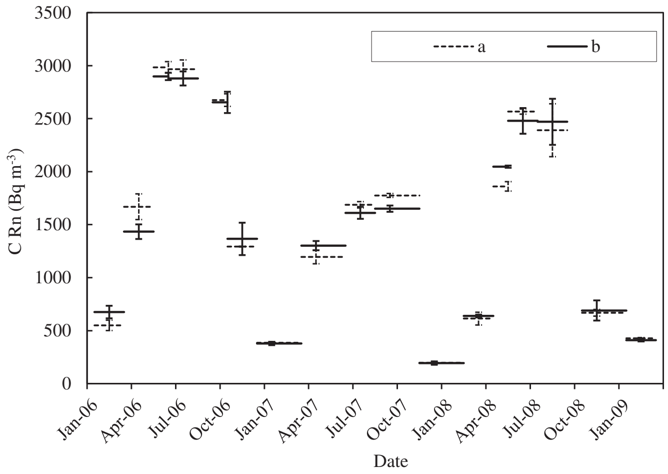


**Fig. 5.** Radon concentrations obtained with the configurations a and b (see Table 1) inside a private basement of the volcanic region of La Garrotxa during three years. Error bars correspond to standard deviation of the mean of two detectors.

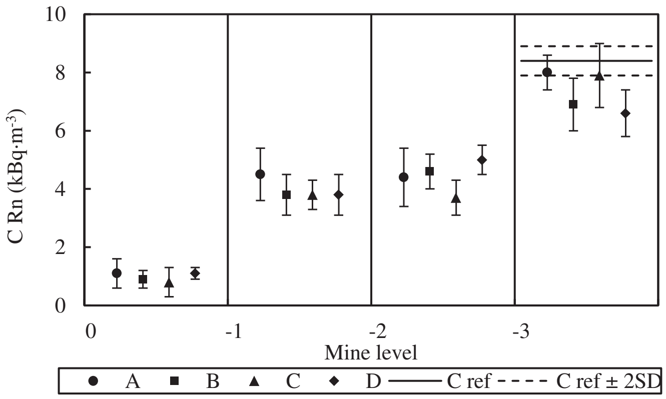


**Fig. 7.** Radon concentration obtained with the configurations A, B, C and D (see Table 1) exposed during 4 days of spring inside underground mines at high humidity conditions and different mine levels. Error bars correspond to standard deviation of the mean of 6 detectors, solid line is the reference value and dashed lines correspond to the standard deviation of the mean ($k = 2$).

inside the INTE radon chamber during the intercomparison exercise. In Fig. 3 we can observe that both configurations present similar behaviour. We obtained a possible temperature effect because sensitivity decreases in exposures at 30 °C (exposures 4 and 6).

In the summer measurement at the underground mines we exposed configurations a and b (Font et al., 2008) and they presented similar results as well (Fig. 4). These first exposures last for one week approximately and we could not already observe the humidity effect over detectors. It was with longer exposures in one of the underground sites with blowhole when we saw the humidity effect over Mylar layer, but its possible impact on track density is hidden by the standard deviation of the results found, so a clear conclusion cannot be stated (Fig. 5). In addition, it also has to be taken into account that samples are small, therefore the sample standard deviation actually is an underestimate of the true standard deviation.

### 3.2. Second phase

In phase II the effect of three different polyethylene bags compared to our reference configuration was analyzed. We have exposed detectors in the INTE radon chamber. The first three exposures have been performed at low humidity conditions and the following two at high humidity conditions (Fig. 6). We have obtained a scattering of the results around 20% so it has not allowed us to see a clear effect. It is worth noting that the exposure time has been around two days, a very short time comparing with the half life of radon, and additionally that after each exposure detectors have been left sealed in the bag for 3 h. There has not been enough time to assure that detectors correctly integrate over varying radon concentrations. Even so, we have observed that the polyethylene bag that seems to less influence detector response is the configuration B, the Tyvek bag.

Under real environmental conditions inside underground ancient mines for a four-day exposure the three polyethylene bags analyzed do not show any clear influence on detectors response because the possible effect is still hidden by the scattering of the results found (Fig. 7). When we increase the exposure time up to more than one week the bag influence seems clearer (Fig. 8) because we have obtained the same behaviour for the three different exposures, and humidity effect is already observable on materials surfaces (Fig. 9). Glass fibre and Mylar layer of dosimeters that were exposed without polyethylene bag (configuration A) present certain damage. If we focus on the configuration B both

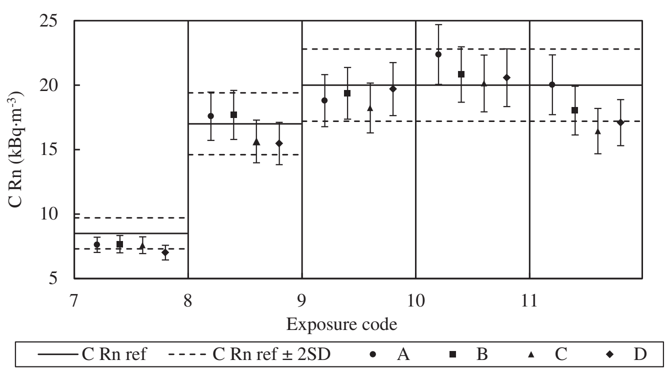


**Fig. 6.** Radon concentration obtained with the configurations A, B, C and D (see Table 1) exposed inside the INTE radon chamber at different controlled environmental conditions. Error bars correspond to standard deviation of the mean of 6 detectors, solid line is the reference value and dashed lines correspond to the standard deviation of the mean ($k = 2$).

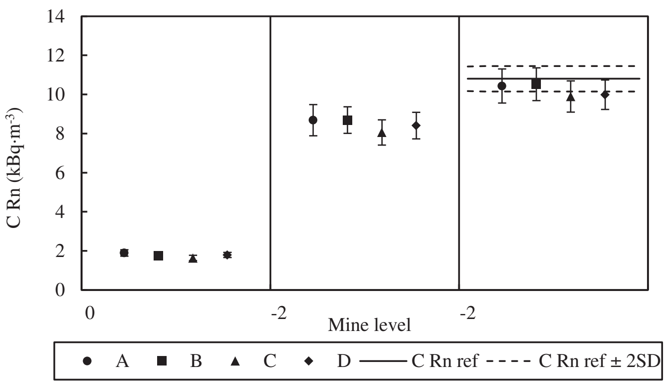


**Fig. 8.** Radon concentration obtained with the configurations A, B, C and D (see Table 1) exposed during the summer inside the underground mines at high humidity conditions and different mine levels: level −2 for 19 days (exposure 3) and 10 days (exposure 4) and level 0 for 29 days (exposure 5). Error bars correspond to standard deviation of the mean of 6 detectors, solid line is the reference value and dashed lines correspond to the standard deviation of the mean ($k = 2$).

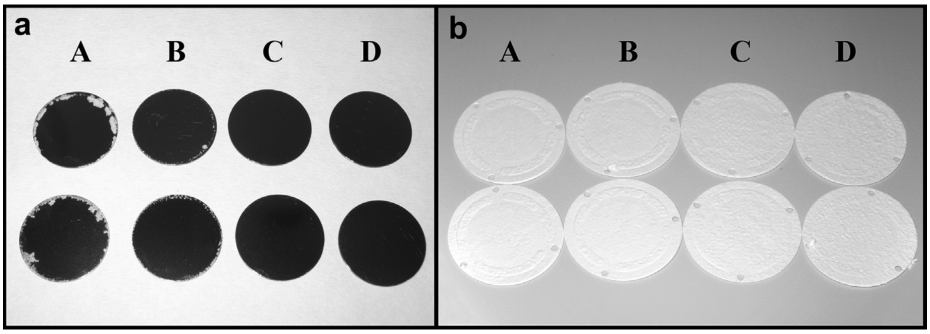


**Fig. 9.** Makrofol detectors exposed with the configurations A, B, C and D (see Table 1) at high humidity conditions during 19 days. Mylar layer (a) and glass fibre filters (b) present certain damages.

start to show signs of deterioration. Therefore, we select configuration D because track density seems not to be affected by this polyethylene bag and it protects enough the detector against humidity for long exposures.

## 4. Conclusions

In some workplaces it might be necessary to carry out long exposures with nuclear track detectors under extreme environmental conditions. We exposed our detectors at different environments with different configurations of polyethylene filters and bags to study the response under high humidity environment.

All filter configurations analyzed have presented similar responses at non extreme conditions (inside a radon box in our laboratory).

Reproducing long exposures at extreme environmental conditions in a controlled radon chamber is not possible for financial and technical reasons. With short exposures in controlled conditions humidity effect was not significant and the polyethylene bag that seems to less influence detector response is the Tyvek bag. But when we use this bag with longer exposures it does not protect enough the detector.

We have to use a real workplace with extreme conditions as a laboratory to perform long-term exposures. Environmental conditions and radon concentrations have to be monitored continuously by means of weather stations and active radon detectors well calibrated.

With one-month exposure we already observed Mylar deterioration due to humidity but the possible impact on track density is hidden by the scattering of the results found. We have observed that the polyethylene bag that seems to less influence detector response and protect enough against humidity for long exposures is the configuration D, the Zipdar bag.

## Acknowledgement

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