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# A step towards accreditation: A robustness test of etching process

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## HIGHLIGHTS

- The evaluation of the robustness of the SSNTD etching process (KOH solution 6.0 N, 75 °C, 270 min) have considered several factors.
- The results evidenced that the etching process can be considered robust.
- The only critical factor is the etching solution's temperature.
- A strict control about stability of temperature during the etching process is needed.

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## ABSTRACT

In the present study the robustness of the etching process used by our laboratory was assessed. The strategy followed was based on the procedure suggested by Youden. Critical factors for the process were estimated using both Lenth's method and Dong's algorithm. The robustness test evidences that particular attention needs to be paid to the control of the etching solution's temperature.



## 1. Introduction

In the framework of the general requirements for the competence of testing and calibration laboratories, the ISO/IEC standard (2005) requires the validation of all non-standard methods. For integrated radon measurements, experimental methods using NRPB/SSI type dosimeters and CR39 plastics (Intercast Europe, Italy) as detectors (SSNTD) can be applied: experimental details about the laboratory-developed method followed in the present work are described elsewhere (Mishra et al., 2005; Orlando et al., 2002).

The validation of a laboratory-developed method is performed to ensure that an analytical methodology is accurate, specific, reproducible and robust over the specified range that an analyte will be analyzed. Some aspects of the quality assurance program for the validation of the integrated radon measurements method were previously described (D'Alessandro et al., 2010). In this work the attention of the authors has been focused principally to the robustness evaluation of the etching process of plastic detectors with the aim to study which factors influencing the final result.

The robustness of an analytical procedure is a measure of its capacity to remain unaffected by small, but deliberate variations in method parameters and provides an indication of its reliability during normal usage. Robustness can be described as the ability to reproduce the analytical method in different laboratories or under different circumstances without the occurrence of unexpected differences in the obtained result.

As well known heavy charged particles, impacting the plastic material surface (SSNTD), cause an extensive ionization of the material that led to the creation of a damaged zone (latent track) along the particles 'path (Nikezic and Yu, 2004).

The etching process allows to visualize the latent tracks and afterward to count them by using optical systems. The performance of the etching process is strictly influenced by the chemical characteristics, the concentration and temperature of the etchant (Hermsdorf et al., 2007). In our method, in order to have easily

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readable tracks (due to radon and its progeny alpha emissions), the following chemical etching conditions are employed: 4.5 h as etching time in a 6.0 N KOH aqueous solution at 75 °C.

The evaluation of the robustness of an etching process is necessary to examine the potential source of variability of quantitative aspect of the method through the variation of variables (inherent to the analytical procedure) called "factors". In particular in this work, to study the main effects of an analytical factor, the "screening design" developed by Plackett and Burman (1946) together with the procedure suggested by Youden (1972) for the robustness evaluation of an experimental method are used.

## 2. Material and methods

As previously mentioned, to study the main effects of an analytical factor, the "screening design" has been used. Screening designs are two-level saturated fractional factorial designs centered on the analytical conditions. Plackett and Burman (1946) developed such design for studying $f$ factors in $N=f+1$ experiments.

The strategy followed to carry out a robustness study is based on the procedure suggested by Youden (1972):

a. identify those factors which can influence the response;
b. for each of these factors define the nominal and extreme levels to be accounted for a routine work, encoding them as follow: nominal value = 0, high value = (+) low value = (−);
c. arrange the experimental plan according to the two-level Plackett and Burman design;
d. perform the experiments in random order and evaluate each factor effect.

In Table 1 together with the real factor to be examined some dummy factors were introduced (factor $b4$, $b7$). The dummy factor is an imaginary factor for which the change from one level to the other has no physical meaning. The dummy factor is necessary to fill in all the columns needed for a Plackett–Burman design with 8 experiments.

To establish the robustness of the SSNTD etching method, 9 experiments were carried out, 8 for the Plackett–Burman design and 1 as a reference. For each experiment 10 SSNTDs, previously exposed to radon atmosphere, were etched. Indeed all radon passive dosimeters, used in the 9 experiments, were exposed to the same radon atmosphere ($1219 \pm 60$ kBqh/m$^3$) in the reference chamber of the Italian National Institute of Metrology of Ionizing Radiation (INMRI-ENEA in the following).

The conditions under which, the experiments were performed, are reported in Table 2: these conditions affect only the etching process of detectors and in particular the tracks' structure.

To determine the influence of the variation of each factor the average track density was determined. Detectors are read out with the Politrack track detector reader, developed at the Politecnico di Milano and supplied by Mi.Am srl (Italy). The reader is an optical microscope with two exchangeable magnifications, about 100 μm and 200 μm, coupled with a 1024 × 768 pixel CCD camera. The spatial resolution is 0.92 μm per pixel for 100 × magnification and 0.57 μm per pixel in the other case. The image is grabbed via firewire by a PC where an image analysis software runs. The same software drives a motorized cartesian Table that moves the detector under the microscope objective (Caresana et al., 2010).

The reading protocol of the laboratory requires that each detector is scanned 10 times. So for each experiment 100 track density values were available. On whole data set statistical analysis (e.g. $t$-test) were performed in order to calculate the average track density for each experiment. In Table 3 the average track density calculated for each experiment is reported. For experiment 0 that is conducted in reference conditions (see Table 1) it is possible to applicate the calibration factor normally used by the laboratory in order to determine the exposure. The exposure so computed is equal to $1198 \pm 120$ kBqh/m$^3$, which is in very good agreement with the one declared by INMRI-ENEA.

**Table 1**
Analytical parameter and relative variations using for evaluating the robustness of the process.

| Factor | | Level | | |
|---|---|---|---|---|
| | | + | − | 0 |
| $b_1$ | Solutions' temperature (°C) | 77 | 73 | 75 |
| $b_2$ | Etching time (min) | 272 | 268 | 270 |
| $b_3$ | KOH concentration (N) | 6.2 | 5.8 | 6.0 |
| $b_4$ | Etching bath | A | B | B |
| $b_5$ | $CH_3COOH$ concentration (N) | 1.1 | 0.9 | 1.0 |
| $b_6$ | Time in $CH_3COOH$ (min) | 12 | 8 | 10 |
| $b_7$ | Number of autofocus | 6 | 4 | 5 |

**Table 2**
Plackett–Burman design for 7 factor ($N=8$).

| Exp no. | $b1$ | $b2$ | $b3$ | $b4$ | $b5$ | $b6$ | $b7$ |
|---|---|---|---|---|---|---|---|
| 1 | + | + | + | − | + | − | − |
| 2 | − | + | + | + | − | + | − |
| 3 | − | − | + | + | + | − | + |
| 4 | + | − | − | + | + | + | - |
| 5 | − | + | − | − | + | + | + |
| 6 | + | − | + | − | − | + | + |
| 7 | + | + | − | + | − | − | + |
| 8 | − | − | − | − | − | − | − |

## 3. Results and discussion

### 3.1. Calculation of effects

For each factor the effect is calculated according to the equation:

$$E_X = \frac{\sum Y(+)}{N/2} - \frac{\sum Y(-)}{N/2} \quad (1)$$

where $X$ represents analytical factor (from $b1$ to $b7$), $E_X$ is the effect of $X$ on the response $Y$ and $\Sigma Y(+)$ and $\Sigma Y(-)$ are the sums of the responses, where $X$ is at the extreme level (+) and (−), respectively, and $N$ is the number of the experiments of the design (8 in our case). For example for $E_{b1,}$ $\Sigma Y(+)$ is given by the sum of the track densities of experiment number 1,4,6,7 and $\Sigma Y(-)$ is given by the sum of the track densities of experiment number 2,3,5,8. The effects can also be normalized respect to the average nominal

**Table 3**
Average density track's for each experiment.

| Exp no. | Track/cm$^2$ |
|---|---|
| 0 | 3490 |
| 1 | 2269 |
| 2 | 3563 |
| 3 | 3511 |
| 4 | 2601 |
| 5 | 3334 |
| 6 | 2724 |
| 7 | 2074 |
| 8 | 3215 |