

## TLD AND ACTIVATED CHARCOAL FOR RADON DOSIMETRY

V. A. Cosma<sup>1</sup>, C. Cosma<sup>2</sup>, I. Pop<sup>3</sup>, V. Benea<sup>2</sup>, Alexandra Flore<sup>1</sup>

<sup>1</sup>Technical University, Automatics and Computers Faculty, 3400 Cluj-Napoca, Ro,  
[porta\\_v@hotmail.com](mailto:porta_v@hotmail.com)

<sup>2</sup>Babes-Bolyai University, Physics Department, 3400 Cluj-Napoca, Ro,  
[amsoc@phys.ubbcluj.ro](mailto:amsoc@phys.ubbcluj.ro)

<sup>3</sup>Technical University, Physics Department, 3400 Cluj-Napoca, Ro

### Abstract

The possibility to combine the track-etched CR-39 or Makrofol detectors with charcoal to increase the detector sensitivity was shown in the last years. Special requests are necessary in the case of a personal dosimeter when the detector must be active only during working activity.

Regarding the combination of charcoal with TL detectors our preliminary investigations indicate that the TLD intensity in normal indoor air ( $\sim 60 \text{ Bq/m}^3$ ) is about two times higher for TLD + charcoal in comparison with simple uncombined TLD. This paper presents a study on the charcoal combined with TLDs and some remarks about charcoal properties. Also, the results show a linear dependence between charcoal TLD signal and radon air concentration.

### Introduction

Radon and radon progeny exposure is the most important source of irradiation for public (1). A lot of investigations have and are being performed in the indoor environment (houses, schools, mines) and also for some workplaces. The most commonly used integrating radon detectors up to now are the activated charcoal and track-etched detectors. The first one is used for short term measurements, typically with an exposure time of a few days and the radon air concentration is found from the radon charcoal content measured by gamma spectrometry (2-4). The second detector is typically exposed for a few months and then chemical and/or electrochemical etched to count the alpha tracks (5). Because of its large adsorption ability, charcoal is often used to collect radon. The principle of the charcoal method is based on the measurement of the gamma emission of radon adsorbed in the charcoal by means of gamma rays detectors such as sodium iodide (NaI(Tl)). Some of those detectors are equipped with a diffusion barrier between the charcoal and the air; therefore the radon adsorbed by the charcoal is proportional to the radon concentration in the air.

The possibility to combine the track-etched detectors CR-39 with charcoal for the growth the detector sensitivity was shown in the last years (6-8). Special requests are necessary in the case of a personal dosimeter when the detector must be active only during working activity. To find the best combination between charcoal and TLD and because each charcoal sort has specific characteristics regarding their adsorption-desorption properties and also different capacities to adsorb the water vapors, the need for a complete relevant study is requested. A lot of data for 20 different sorts of activated charcoal were presented by Scarpitta (9-10), as a study in the radon chamber for 2.8 days at 14% relative humidity (RH) using open-faced canisters of 1-1.5 cm thickness of charcoal bed. For some of these charcoals having better characteristics for radon adsorption were also made measurements for 43%, 51%, and 70% RH. The response of charcoal to humidity variations using both diffusion and convection of radon also influence of the desiccant on radon adsorption has been investigated (11).

TLDs have only a limited utilization in radon dosimetry because they are generally sensible to whole spectrum of natural radiation: cosmic, terrestrial or gamma-beta radioactivity of building materials. But they can be used for radon progenies (RP) measurement. The TLD type contains an air-sampling pump that draws a continuous, uniform flow of air through a detector assembly. The detector assembly includes a filter and at least two TLDs. One TLD measures the radiation emitted from radon decay products collected on the filter, and the other TLD is used for a background gamma correction. This RP is intended for a sampling period of 48 hours to a few weeks. Analysis of the TLD is performed in a laboratory using a TLD reader. Interpretation of the results of this measurement requires a calibration for the detector and the analysis system based on exposures to known concentrations of radon decay products (12).

Stranden (13) shown that charcoal detectors can be equipped with a TLD chip inserted into the charcoal, the radon concentration is then determined using a TLD reader. Those detectors are convenient for the determination of indoor radon concentrations because of their low cost, the short time of exposure required and their small size.

The main characteristics involved in the design of a combined Charcoal-TLD detector are: the background radioactivity of the charcoal, the adsorption and desorption time, directly connected with thickness of charcoal bed, the  $k$  value of adsorption coefficient and the breakpoint of the charcoal which is directly related with the water mass in charcoal. In the case of an important quantity of radon adsorbed in charcoal, the achievement of this breakpoint leads to the substitution of radon with water vapors, diminishing drastically the adsorbed radon by charcoal (9). Recently, Scarpitta, (14) made a comparative study using the best known charcoal, Calgon PCB, and a new charcoal sort, Carboxen 564, produced by Supelco Inc, Belafonte, USA. It was found that the last have superior characteristics regarding the adsorption properties of the radon especially in thin layers (15). Using this charcoal and Makrofol track detectors a radon dosimeter for work places exposure was proposed by A. van Deynse (16).

Very recently studies about a TLD-Charcoal dosimeter were made by a Polish team (17-18), which used a very sensitive TLD chip, namely MCP-N (LiF: Mg, Cu, P), produced in their laboratory.

In the present work we will present our preliminary results obtained using the above-specified TLD chips (LiF: Mg, Cu, P) and a sort of Romanian charcoal of which properties were studied in radon laboratory from Ghent University (8,15,16).

In the future we intend to use these sensitive chips coupled with a very special sort of charcoal mentioned above, Carboxen 564, to improve the detection limit and to obtain a personal radon dosimeter possible to be used for work place exposures.

## Background theory

For the equilibrium adsorption of radon by charcoal (4) one can write:

$$A = k \rho C \quad (1)$$

where  $A$  and  $C$  are respectively the activity concentrations of radon in the charcoal and in the air ( $\text{Bq/m}^3$ ),  $k$  is the adsorption coefficient ( $\text{m}^3/\text{kg}$ ) and  $\rho$  is the bulk density of charcoal ( $\text{kg/m}^3$ ). From the data reported in the literature ( $\rho \approx 500 \text{kg/m}^3$  and  $k \approx 4 \text{m}^3/\text{kg}$ ) it can calculate that the radon concentration in the charcoal is approximately three orders of magnitude higher than in the air. For practical use of a dosimeter based on the combined TLD-charcoal technique the time of adsorption and desorption from the charcoal bed is a very important parameter. This constant can be measured and a strong dependence of the thickness of the charcoal bed was observed (9-10). The quickness of adsorption is characterized by build-up time constant (BUTC) and can be determined from:

$$\text{BUTC} = t_s / \ln(k_{\max} / (k_{\max} - k(t_s))) \quad (2)$$

where  $k(t_s)$  is the dependence of adsorption coefficient  $k$  of the adsorption time,  $k_{\max}$  being the equilibrium  $k$  coefficient utilized in equation (1) and  $t_s$  is the time required to achieve 67% of  $k_{\max}$ .

The desorption is characterized by desorption time constant (DTC) related of the  $t_{1/2}$  time (the desorption of a half radon from charcoal) as:

$$\text{DTC} = t_{1/2} / \ln 2 \quad (3)$$

These two constants (BUTC and DTC) measure "the radon-charcoal" inertia, being very important parameters for designing a simple dosimeter or a personal dosimeter.

An important characteristic of charcoal is the breakpoint (14). The breakpoint represents the mass of adsorbed water vapors which if is surpassed the water molecules substitute the radon atoms from the inner structure of charcoal. As results the  $k$  coefficient drastically decreases. The background alpha radioactivity of charcoals also can influence the signal induced in TLD chip, especially at low radon concentrations and for long time exposure.

## Experimental Method

The radon dosimeter is based on radon adsorption on activated charcoal. By placing TLD crystals inside an open charcoal container, the beta and gamma radiation from radon daughters, produced by the decay of adsorbed radon, is detected continuously during the exposure time (13).

The adsorption constant also the background radioactivity of the different charcoals was measured by gamma spectrometry using a Ge calibrated detector. For the radon evidence the main two photo peaks of the radon progeny (609 keV and 352 keV) were used. Radon concentration in the air was measured using a Radim device (18). The sample Nr. 7 (Ro-2F, from Table 1) was measured in Ghent and remeasured in our laboratory. It is a Romanian sort of charcoal (CASS-Buzau) and this sort was used in our experiments. Because for low radon concentration the charcoal radioactivity itself can contribute to the TLD signal we presented in Table 2 the background radioactivity of different

types of charcoal. A special device for rapid and complete charcoal cleaning (complete activation) used by us is shown in Fig.1. A brass cylindrical vessel of 0.8 l is connected to a vacuum pump and in this time the temperature is maintained at 100°C (boiling water). After 15-20 min of pumping the charcoal is removed and its radioactivity after 3-4 h (radon progeny disintegration) the background activity of charcoal is obtained.

The Polish TLD chips, namely MCP-N (LiF: Mg, Cu, P) type, 3 mm in diameter and 1.2 mm thick were installed in the center of two kinds of charcoal plastic canisters of 10 ml and 30 ml in volume. These plastic canisters have many holes symmetrically delivered to allow easy entrance of radon gas. The canisters were exposed typically for 120 h in a radon chamber (58 l) at 4 different concentrations: 850 Bq/m<sup>3</sup>; 1240 Bq/m<sup>3</sup>; 1970 Bq/m<sup>3</sup> and respectively 3260 Bq/m<sup>3</sup>, Fig.2. These radon concentrations were continuously measured with the Radim monitor device inside (18). After exposure, the reading of TLD was made with Harshaw -2000 device.

Fig.1. The device for charcoal degassing.

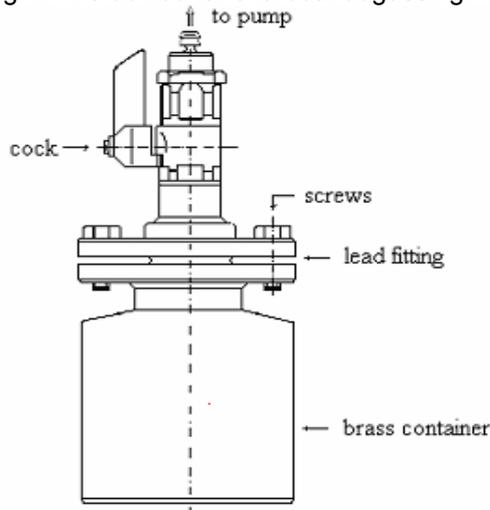
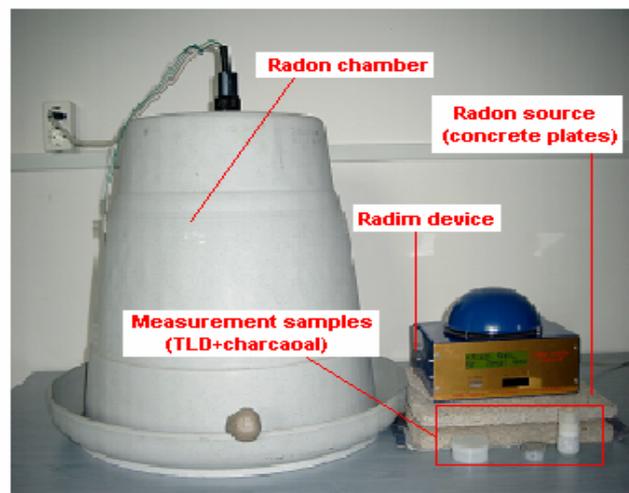


Fig. 2. The radon chamber and Radim device



### Results and discussion

In Table 1 are shown the adsorption constants for eleven charcoal sorts measured at Gent University laboratory (8). Carboxen charcoal has the best adsorption properties. In the table is marked with bold letters the charcoal used by us (Nr.7), in our preliminary measurements. By using Carboxen 564 instead of Ro-2F the sensibility of method can increase about five times.

Table 1. The results of exposure at low radon concentration (120 Bq/m<sup>3</sup>) at 60%RH (8)

Nr. crt.	Type of charcoal	Nr. samples	Gamma activity (counts/g)	k (m <sup>3</sup> /Kg)
1	Ro-1G	2	83	4.02
2	George	4	56(15%)	2.75
3	CalgonBPL	3	56(16%)	2.75
4	CalgonPCB	3	75(10%)	3.61
5	Norit -R1	4	61(12%)	2.9
6	PZ	1	25	1.52
7	<b>Ro-2F</b>	<b>4</b>	<b>20(20%)</b>	<b>1.42*</b>
8	Merck	1	21	1.05
9	ART2515	1	19	0.98
10	Carboxen (2 mm)	5	140(8%)	6.92
11	Carboxen (3 mm)	5	148(7%)	7.1

\*Remeasured in our laboratory

In Table 2 one can see the background radioactivity of some charcoals measured with a Ge high resolution spectrometer after a very good degassing and kept in old air for two weeks. Carboxen 564

has a very low background closed of Ge background (last row) whereas our charcoal Ro-2F has the background with 10% higher, especially due to radium and cesium content.

Table 2. Gamma background of different charcoals.

Type	$E_\gamma > 90 \text{ keV}$		Photopeak area		Obs.
	(A)	(B)	(609 keV)	(352 keV)	
CalgonPCB	-	107976	48	92	Low activity
George	101380	100800	49	52	Background
Norit R1	112600	109263	180	250	$^{226}\text{Ra}$ , $^{137}\text{Cs}$
Ro-1G	121280	117500	233	346	Much $^{226}\text{Ra}$
<b>Ro-2F</b>	<b>109700</b>	<b>110400</b>	<b>161</b>	<b>201</b>	$^{137}\text{Cs}$ , $^{226}\text{Ra}$
Merck	-	104200	-	-	Low activity
PZ	-	108300	-	-	$^{137}\text{Cs}$
Carboxen	100700	100600	58	80	Low activity
Ge backgr.	100700	100600	30	54	

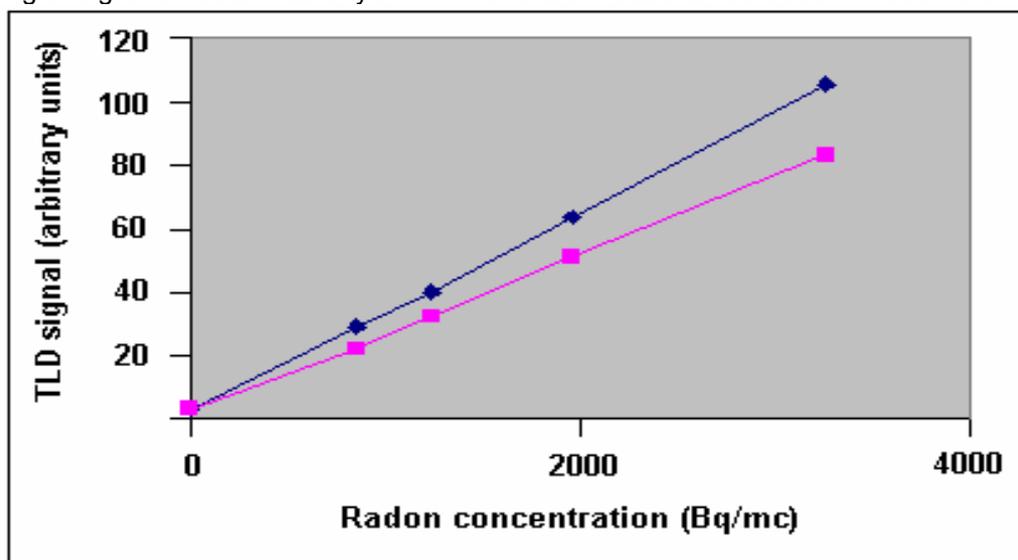
The results for 4 chips simultaneous exposed in the radon chamber at  $1240 \text{ Bq/m}^3$  (5 days) in  $V_1$  (30 ml) and  $V_2$  (10 ml) canisters are shown in Table 3. The TLD intensity of background was for these chips  $B=4\eta C$ . As can see, the reproducibility is better as 10%. If we consider in  $V_1$  case the detection limit as  $B^{1/2}=2\eta C$  the minimum detectable concentration for one day exposure is about  $400 \text{ Bq/m}^3$ . The result recently obtained by Bogacz *et al.* (17) is  $100 \text{ Bq/m}^3$  for three days of exposure. Considering usage of Carboxen 564 the limit of detection can be five times smaller, therefore  $80 \text{ Bq/m}^3/\text{day}$ , rather small and promising to be used for a radon personal dosimeter at workplaces (16). Another experiment was the exposure of 3 TLD + charcoal and others 3 TLD only chips in the laboratory room (about  $60 \text{ Bq/m}^3$ ) for 5 days. The average corrected background signal was two times higher in the case of TLD + charcoal sets.

In Fig.3 are presented the results obtained for the five days exposure of TLD + charcoal in the radon chamber at four different concentrations. A linear dependence between the signal intensity and radon content in the radon chamber can be seen both for  $V_1$  and  $V_2$  sample types as in paper (17). Although  $V_1$  canister is three times larger as  $V_2$  we can see from Table 3 and Fig.3 that the intensity of the signal is only with 27% higher. Probably the optimum canister volume is about 40-45 ml therefore about 20-25 g.

Table 3. Four TLD + charcoal exposures at  $1240 \text{ Bq/m}^3$  for 5 days

Chip number		$P_1$	$P_2$	$P_3$	$P_4$	Mean	Standard deviation
TLD signal	" $V_1$ "( $\eta C$ )	33.7	32	29.1	29.7	31.16	2.12
	" $V_2$ "( $\eta C$ )	24.1	27.2	25.3	21.7	24.57	2.29

Fig.3. Signal emission intensity of TLD + charcoal versus concentration in radon chamber



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