Continuous measurement system of radon concentration in water by gamma radiation detection emitted by ^{214}Bi and ^{214}Pb decay (*)(**)

G. GALLI, C. GUADONI and C. MANCINI

Università di Roma "La Sapienza", Dipartimento di Energia Nucleare e Conversioni di Energia Roma, Italy

(ricevuto il 9 Giugno 1998; approvato il 10 Febbraio 1999)

Summary. — Systematical and continuous measures of ^{222}Rn concentration in groundwater have shown a possible correlation between ^{222}Rn anomalies and seismic events. The system proposed in this work will draw water from a source, fill a 0.69 l beaker Marinelli (COUNTESS R. J., *Measurement of* ^{222}Rn *in water, Health Phys.*, **34** (1978) 390-391) and detect main γ lines emitted from ^{222}Rn by using a $2'' \times 2''$ NaI(Tl) scintillator surrounded by a low background γ spectrometer (DE LUCA A. and MANCINI C., *The measurement system for* ^{222}Rn *monitoring with charcoal adsorption collectors, Health Phys.*, **61** (1991) 543-546).

PACS 91.10.Tq – Earth tides.
PACS 91.25.Ey – Interactions between exterior sources and interior properties.
PACS 92.40.Kf – Groundwater.
PACS 01.30.Cc – Conference proceedings.

1. – Introduction

Certain experimental evidences, particularly those collected in occasion of the Kobe earthquake [1] on 17 January 1995, have shown a possible correlation between the anomalies of ^{222}Rn concentration in groundwater and seismic events [2-5]. In order to verify the existence of such correlation it is necessary to perform systematical and continuous measures of ^{222}Rn concentration in groundwater in areas frequently interested by seismic events, so as to collect significant data. In this way, we have realized the system described in this work, that will be installed in a deep well located in Frascati, in the area of Castelli Romani characterized by a significant seismic activity. Normally, ^{222}Rn concentration in the water of the selected well is about 200 Bq/l.

(*) Paper presented at the "Fourth International Conference on Rare Gas Geochemistry", Rome, October 8-10, 1997.
(**) The authors of this paper have agreed to not receive the proofs for correction.

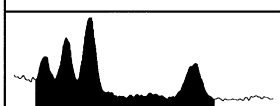
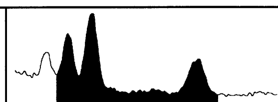
2. – The methods of measurement

Methods used to measure ^{222}Rn concentration in water are based on the detection of the radiation emitted by the decay of ^{222}Rn itself and of its short-lived daughters. Measures are performed detecting alpha-particles emitted by ^{222}Rn, ^{218}Po and ^{214}Po, or the gamma-rays emitted by ^{214}Pb and ^{214}Bi.

The most employed system uses scintillation flasks, called Lucas Cell [6], coupled with a photomultiplier tube. The scintillation induced by alpha-particles on a thin layer of ZnS(Ag) that covers the internal surface of the cells is detected. The pulse spectrum obtained using scintillation cells is a continuous spectrum and it is not possible to correlate events with energies of alpha-particles; therefore this system consists of a counter that records the overall number of counts in the selected time interval and it is not possible to have a continuous check of the good correlation between counting rate and ^{222}Rn concentration.

The system proposed in this work is based on the detection of the most abundant gamma radiations emitted by ^{214}Pb (main energies 242, 295 and 352 keV) and ^{214}Bi (main energy 609 keV). During the monitoring of ^{222}Rn concentration, the analisys of the spectrum of each measure allows to check the reliability of the collected data. Upon the spectrum it is possible to integrate counts over different ROIs (Regions Of Interest). Table I shows a comparison between two NaI(Tl) scintillators, respectively $2'' \times 2''$ and $3'' \times 3''$, for different ROIs shown above each column: it is remarkable how the $S/\sqrt{B}$ (S: overall counts, B: background counts) is nearly the same for a given ROI, while MDA (Minimum Detectable Activity) results to be about 2 Bq/l for the $3'' \times 3''$ scintillator and comprised between 4 Bq/l and 5 Bq/l for the $2'' \times 2''$ one when a 15 min counting time is concerned [7].

TABLE I. – *A comparison between* $2'' \times 2''$ *and* $3'' \times 3''$ NaI(Tl) *scintillators used in a low background* γ*-ray spectrometer.*

	Region Of Interest					
						
	2" × 2"	3" × 3"	2" × 2"	3" × 3"	2" × 2"	3" × 3"
Counting efficiency (count/Bq)	67,8 ± 2,3	112,4 ± 4,2	54,9 ± 1,9	90,1 ± 3,4	46,2 ± 1,6	75,2 ± 2,53
Signal to Noise Ratio	228,5 ± 1,1	245,3 ± 2,7	202,6 ± 1,1	218,7 ± 2,7	223,1 ± 1,2	249,3 ± 4,5
LLD (count/min)	13,05 ± 0,01	14,42 ± 0,10	11,92 ± 0,01	12,97 ± 0,10	9,11 ± 0,01	9,50 ± 0,10
MDA (Bq/l)	4,19 ± 0,14	2,03 ± 0,08	4,72 ± 0,16	2,27 ± 0,09	4,29 ± 0,15	1,99 ± 0,07

Since the system realized is appointed to monitor ^{222}Rn concentration of the order of one hundred Bq/l, we have chosen to work with the $2'' \times 2''$ scintillator due to its smaller dimension and lower price. Through a usual counting chain, equipped with a preamplifier and an amplifier, the detector is connected to a multichannel analyzer.

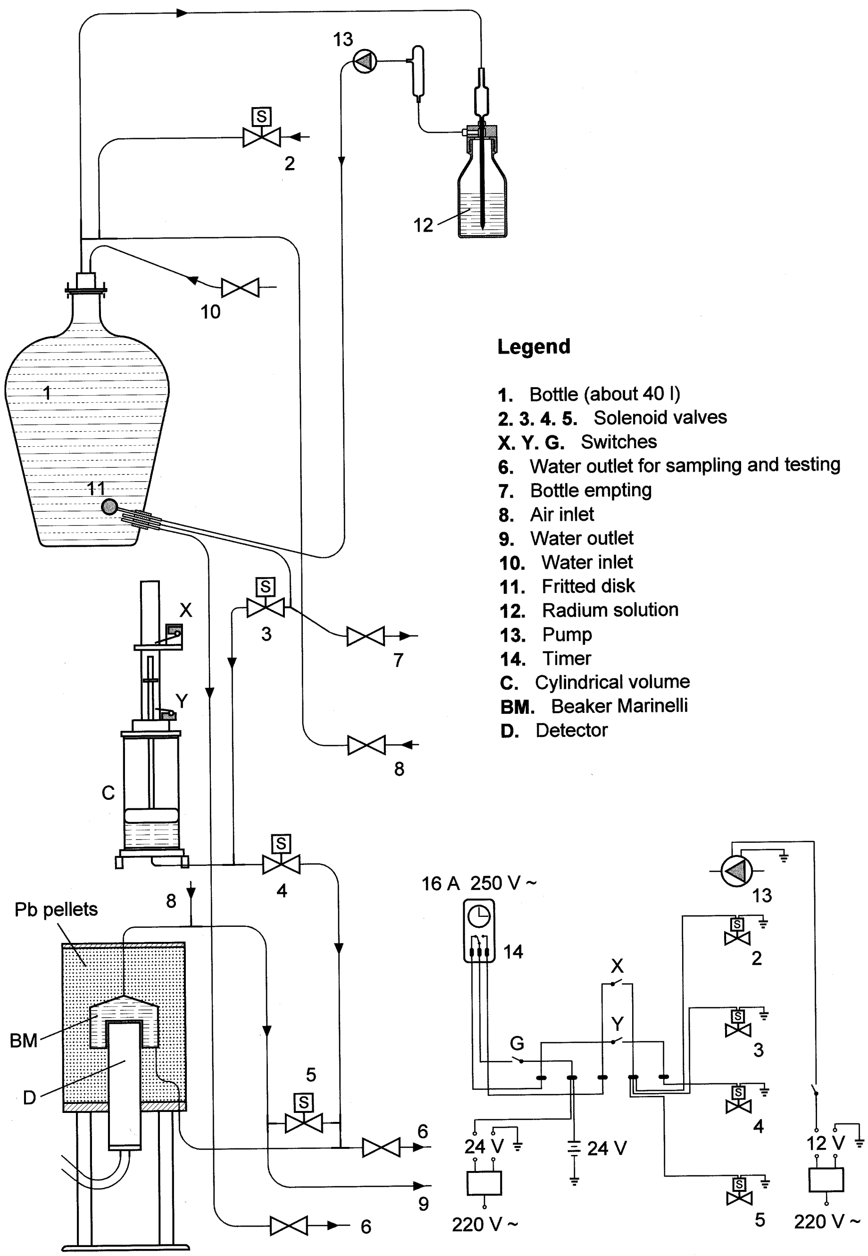


Fig. 1. – Hydraulic and electrical schemes of the laboratory test facility.

The shield needed to reduce the background contribute consists of lead marbles, which allows an easy carrying. Lead thickness is 50 g/cm^2.

Measures can be performed continuously with a constant water flow through the Marinelli beaker. In this case it is necessary to provide a tank of sufficient volume between the well and the system so as to ensure the establishment of radioactive equilibrium between ^{222}Rn and its short-lived daughters before the measure, *i.e.* at least 3 hrs. This operation mode needs a continuous working of the well pumps and involves a large consume of water, not always acceptable. Due to this consideration and because of the characteristic times of the phenomena investigated, we have chosen to do one measure every four hours, with a 15′ counting three hours and a half after the water sample has been taken.

3. – Laboratory tests

Before setting up the system by the well, some laboratory tests were carried out using the facility shown in fig. 1, where a 40 l bottle simulates the water spring. The bottle is filled through the valve 10. Pump 13 is swiched on for about 30 minutes and leads to a recirculation of the residual air, which bubbles in the bottle and in a flask partially filled with a $RaCl_2$ solution. ^{222}Rn extracted from solution is partially transferred to the water in the bottle. Using a 30 kBq source we have obtained a ^{222}Rn concentration in water of about 300 Bq/l at normal pressure and temperature.

According to the electrical scheme shown in fig. 1, a timer controls the solenoid valves 2, 3, 4 and 5. Valves 2, 3 and 5 are operated in parallel: the first one allows the filling-up of the cilynder C, provided with a floating piston. A connecting rod moves

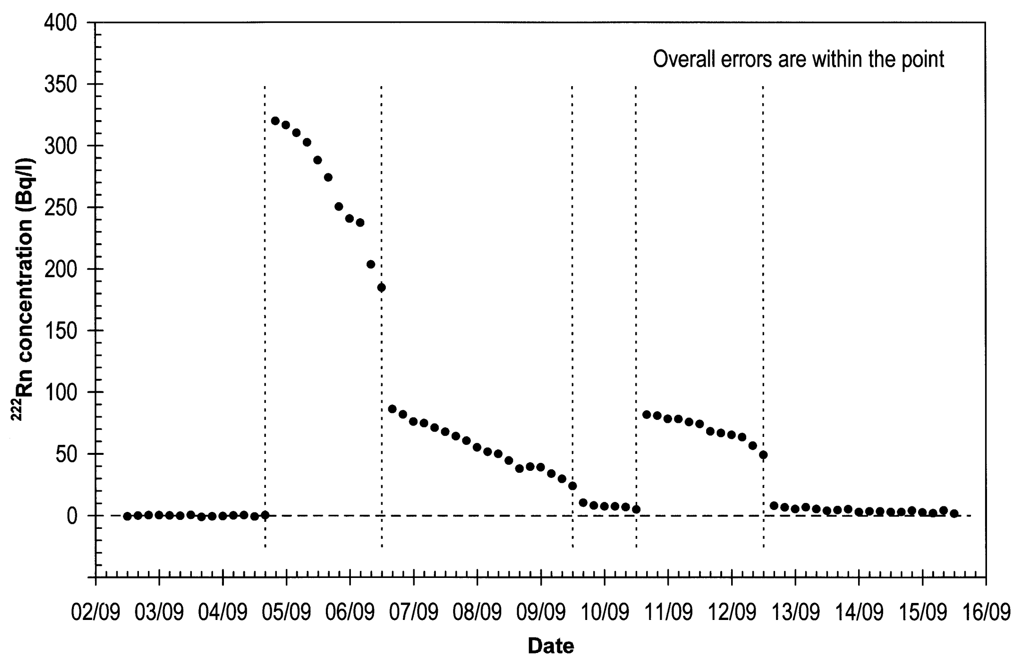


Fig. 2 – Response *vs.* time to variations in ^{222}Rn concentration.

jointly with the piston and actuates two switches X and Y, respectively when the cylinder is full and empty. Valve 2 lets the air enter the bottle during the transfer phase.

Each complete cycle lasts 4 hrs. At the beginning, the timer changes over for 15′ actuating valves 2, 3 and 5 until the floating piston reaches within a few minutes the switch X which stops the water flow. Valve 5 allows the emptying of the Marinelli beaker which contains the water analyzed in the previous measure. When the first 15′ have elapsed, the timer changes over opening the valve 4 which allows the emptying of cylinder C through the beaker. The cylinder volume is about triple with respect to the beaker volume (0.7 l) to ensure that the water to be measured has not come into contact with air.

At the end of the water transfer the switch Y de-excites valve 4. About 3 h and 15′ later the MCA starts counting for 15′ and stops before the next cycle starts.

^{222}Rn concentration behaviour relative to data collected over a 14 days' test is shown in fig. 2. The first region refers to water remained in the airtight bottle for almost a month and data collected can be regarded as background. The next region is subsequent to an enrichment cycle.

After 44 h we added mineral water to dilute the enriched water in the bottle. After a further diluition, on 10/9 at 12.00 we proceeded to a smaller enrichment.

The test has been concluded emptying completely and filling again the bottle with water from the municipal aqueduct.

The system stability has been verified comparing spectra from a three days' test.

4. – Conclusions

Within 1997 the system, limited to a spectrometer, will be installed by a well in Frascati. The well pump will be run for a few minutes at the beginning of each cycle and water will flow through the Marinelli beaker providing a complete renewal. The realized facility will be used for tests with other systems in progress.

REFERENCES

[1] IGARASHI G. *et al.*, *Groundwater radon anomaly before the Kobe earthquake in Japan, Science*, **269** (1995) 60-61.

[2] FRIEDMANN H. and HERNEGGER F., *A method for continuous measurement of radon in water of spring for earthquake prediction, Geophys. Res. Lett.*, **5** (1978) 565-568.

[3] FRIEDMANN H., *Anomalies in the radon content of spring water as earthquake precursors phenomena, Earth. Pred. Res.*, **1** (1985) 179-189.

[4] HAUKSSON E., *Radon content of groundwater as an earthquake precursor: evaluation of worldwide data and physical basis, J. Geophys. Res.*, **86** (1981) 9397-9410.

[5] IGARASHI G., TOHJIMA Y. and WAKITA H., *Time-variable response characteristics of groundwater radon to earthquakes, Geophys. Res. Lett.*, **20** (1993) 1807-1810.

[6] LUCAS H. F., *Improved low level alpha scintillation counter for radon, Rev. Sci. Instrum.*, **28** (1957) 680.

[7] BERNARDINI L., *Misura in continuo della concentrazione di radon in acqua*, Tesi di Laurea, Dipartimento di Energia Nucleare e Conversioni di Energia, Università di Roma "La Sapienza" (1997).