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Introduction

In the last decades an increasing attention has been given to the problem of reducing the radon in several environments.

This problem is particularly evident in mine environments and in those places with a limited air flow-rate, where rocks or building materials might emanate remarkable radon amounts.

The solution usually adopted in this context is to assume an adequate hourly-exchange of fresh air in order to achieve a "normal" radon level of about 30 Bq/m^3 in the considered environment.

The simple solution mentioned above is not satisfactory for particular types of experiments in which an extremely low background of radioactivity is needed; in these experiments researchers aim to obtain radon levels orders of magnitude lower than the environmental radon values.

As well as the experiments on solar neutrino {*KAMIOKANDE* [Ref. 1], *GALLEX* [Ref. 2], *SNO* [Ref. 3]}, also the experiments on *Dark Matter* and on the $\beta\beta$ -decay are directly interested in the attainment of such a low level in radon contamination.

As regards the installation of the *BOREXINO* prototype (Counting Test Facility) in the Hall C of the underground laboratory of the LNGS, the problem consists in the reduction of the value of the radon concentration in a very large volume 1000 m^3 down to a level less than 0.1 Bq/m^3 .

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Working principles of radon filters for gases

The so-called radon filters are not “filters” in the usual sense of the word. A strict definition of a filter would describe it as a device which does not allow the passing through of some components of a given mixture. When a gas containing radon (dry air, for instance) is in contact with a filter (such as a charcoal bed) the radon atoms spend a long time in contact with the filter material, but are not permanently adsorbed by it. Inside a “filter” device, in this case, a radon molecule continues to be alternatively adsorbed on the filter (for a length of time of mean value t_f) and desorbed from it into the surrounding gas (for a mean time t_g), being $t_g \ll t_f$.

Let's assume to have a gas of volume V_g in contact with a given filter material of mass m_f , density d_f and volume V_f . In a state of thermodynamical equilibrium, the number of molecules leaving a surface S of the filter material is equal to the number of adsorbed ones; when any saturation of the capturing sites and the competition with other kinds of molecules, such as water vapours, is negligible (§), the following relationship can be written:

$$\frac{N_g}{V_g t_g} = \frac{N_f}{V_f t_f} \quad \{1\}$$

The ratio:

$$K = \frac{N_f/m_f}{N_g/V_g} \quad \{2\}$$

is said to be the *equilibrium constant* of the process. This value is usually measured in m^3/kg and it strongly depends on the temperature. A list of K values at ambient temperature

(§) Our real interest is the radon removal from dry air or from dry nitrogen. In these two cases the assumptions done above are well verified. A detailed study of a more complicated situation of practical interest for people using charcoal canisters in order to determine the radon distribution in different possible environments is given in (1).

for different charcoal types is given in [2]. The law of Dalton and Henry applied to equilibrium between gases and solids would give a straight exponential dependence upon temperature. This is confirmed by experimental data, at least in a limited range of temperatures around room temperature [3,4]. A high adsorption charcoal has a typical K of $4.3 \text{ m}^3/\text{kg}$ at $20 \text{ }^\circ\text{C}$ and a K of $12 \text{ m}^3/\text{kg}$ at $0 \text{ }^\circ\text{C}$.

From {1} and {2} we can deduce:

$$\frac{t_f}{t_g} = K d_f \quad \{3\}$$

Assuming the already given values for K and a density of the order of 1000 kg/m^3 , it is easy to verify that in a good filter, such as charcoal, t_f is really much greater than t_g .

It may be helpful to note that {1} can be rewritten as:

$$C_f = \frac{n_f}{V_f} = \frac{K d_f P_{Rn}}{RT} = \frac{K d_f P X_{Rn}}{RT} \quad \{4\}$$

where C_f is the concentration of the radon adsorbed on the charcoal, n_f is the number of adsorbed radon moles, P_{Rn} is the partial pressure of the radon gas, X_{Rn} the radon molar fraction in the gas, P the total gas pressure.

Static filters

It is often convenient to rewrite {2} as:

$$N_f = K m_f \frac{N_g}{V_g} \quad \{5\}$$

This equation constitutes the working base of a very widespread and cheap radon measuring instrument [5].

It consists of a canister containing a given amount of charcoal (about 0.1 kg) which is exposed to the air of a given site, after it has been flushed using hot air. After a few days of

exposition, the equilibrium condition correspondent to the relationship expressed by equation {5} should be achieved and a straightforward measurement of the activity of the decay products of the N_f nuclei of Rn will permit to deduce the value of the ratio N_g/V_g , that is the radon concentration in the air of the exposition site.

The canisters briefly described could be thought as prototypes of possible static radon filters; they can be used in order to reduce the radon concentration in a close volume which initially contains N_o radon atoms. At equilibrium the radon atoms will be partly dissolved in a gas volume V_g and partly adsorbed on a mass m_f of charcoal. The initial radon concentration N_o / V_g will be reduced to the value $(N_o - N_f)$, where N_f is given by:

$$\frac{N_f}{N_o} = \frac{Km_f}{Km_f + V_g} \quad \{6\}$$

Assuming that the gas is really dry and does not interact with the charcoal, the main problem with such a type of filters is that the diffusion process is quite slow (few days), which means that comparatively long times are required to reach an equilibrium concentration.

The retention time of fluxed filters

In order to reach the equilibrium condition {6} in a faster way, it is possible to flux the gas through the filter. In this configuration, a volume V_g of gas is continuously fluxed through a cylinder of cross section S and length L ; the cylinder contains a mass m_f of "filter" material. It is assumed that at the beginning the filter material does not contain radon at all and u is defined as the mean velocity of the gas while it flows between the filter granules. As soon as the radon atoms begin to flow, they will almost immediately be captured by the charcoal; after a mean time t_f , they will be desorbed for a time t_g and will drift with the gas at velocity u before being adsorbed again. In the described process the

radon molecules will not be permanently fixed in the filter material; the filter action will only consist in slowing down their drift motion.

Therefore it is possible to define the "retention time" T_r of the filter (+), which is equal to the mean time required for a radon molecule to pass through the filter.

As regards the cylinder under consideration, the retention time is given by:

$$T_r = \frac{L(t_f + t_g)}{u t_g} \approx \frac{L t_f}{u t_g} = \frac{L K d_f}{u} \quad \{7\}$$

(assuming a typical velocity of 0.2 m/s and a cylinder length $L=0.1 \text{ m}$, T_r turns out to be many thousands of seconds).

Obviously the individual crossing-time of different molecules will be different and will be distributed around the average mean value computed above. The proper dispersion can be evaluated and taken into account {6,7}.

Possible ways of controlling the radon concentration

All the concepts needed to discuss the behaviour of the radon concentration in a closed vessel of volume V_g containing dry gas which is continuously fluxed through a charcoal filter have been introduced.

We will give the following notations:

- E_m number of molecules per second entering the gas desorbing from the vessel walls
- E_f number of molecules per second entering the gas desorbing from the filter material as a consequence of a radioactive decay of uranium or thorium atoms contaminating the filter material
- E_i number of molecules per second mixing with the gas entering from any other possible source (molecules desorbed from various objects inside the vessel, entering the vessel through holes in the vessel walls, etc)

(+) The existence of the retention time is the basis of the long time established techniques of gas chromatography.

- T^* time required to flux a volume V_g of gas through the filter
- $N_f(t)$ number of radon molecules that are inside the filter at the time t
- $N_g(t)$ number of radon molecules in the gas outside the filter at the time t
- C_g radon concentration in the gas ($C_g = N_g / V_g$)

Given these notations, it is easy to write a simple equation to describe the change of N_g with time; the relationship is given by:

$$\frac{dN_g(t)}{dt} = (E_m + E_f + E_i) - \frac{N_g(t)}{\tau} - \frac{N_g(t)Su}{V_g} + \frac{N_g(t-T_r)Su}{V_g} e^{-T_r/\tau}$$

Introducing $T^* = V_g / S \cdot u$:

$$\frac{dN_g(t)}{dt} = (E_m + E_f + E_i) - \frac{N_g(t)}{\tau} - \frac{N_g(t)}{T^*} + \frac{N_g(t-T_r)}{T^*} e^{-T_r/\tau} \quad \{8\}$$

The first term on the right hand side describes the "radon sources"; the second term the effect of natural radioactive decay; the third term is due to the capture of radon molecules into the filter; the fourth term represents the appearance at the filter output of other radon molecules which passed the filter without decaying.

When an equilibrium concentration is established (after a few T^*), the above equation becomes:

$$0 = (E_m + E_f + E_i) - \frac{N_g}{\tau} - \frac{N_g(1 - e^{-T_r/\tau})}{T^*} \quad \{9\}$$

The most important result to be pointed out is that a good filtering action of the radon can be practically achieved only if $T^* \ll \tau$ and $T_r \gg \tau$. In that case the relationship {9} can be simplified as:

$$0 = (E_m + E_f + E_i) - \frac{N_g}{T^*} \quad \{10\}$$

which demonstrates that the equilibrium radon concentration is:

$$C_g = \frac{N_g}{V_g} = (E_m + E_f + E_i) \frac{T^*}{V_g} \quad \{11\}$$

This value has to be compared with the concentration C_{0g} without any filter:

$$C_{0g} = (E_m + E_f + E_i) \frac{\tau}{V_g} \quad \{12\}$$

The comparison demonstrates that the use of a fluxed filter, under the best experimental conditions, can reduce the natural radon concentration only down to a factor τ / T^* . The value of the ratio τ / T^* depends on the volume of the gas that has to be purified and on the air flow-rate through the filter. The most difficult radon isotope to filter is the comparatively long lived ^{222}Rn whose half-life is ≈ 91 h. According to the equations listed above, in a vessel of volume equal to V cubic meters, a reduction of two order of magnitude of the radon concentration will require a filter able to purge about V cubic meters of air per hour.

Possible practical solutions

The discussion now will be focussed on the possibility of filtering the comparatively long lived (3.82 d = 91 h) ^{222}Rn atoms from air.

The first possibility is to make the retention time in the filter longer than the isotope mean life: in that case most of the radon nuclei entering the filter will decay before reaching the filter end. The solution is conceptually simple but it is not so easy to realise; particularly, if the filter has to operate at

room temperature (at 293 K), with $u = 0.1 \text{ m/s}$, the minimum filter length L will be greater than *20 meters* !!

According to the {4}, the filter length notably decreases on reducing the temperature; anyhow, it's practically impossible to achieve thin filters; for instance, assuming a reasonable exponential correlation between the equilibrium constant K and the temperature, the minimum filter length at 200 K turns out to be greater than *1 m*.

The second possible solution will consist in the alternate use of two identical filters, characterised by a retention time much shorter than the isotope mean life.

The basic ideas underlying this scheme are the following ones:

- while one of the filter is in use, the other filter is being purged, in order to desorb the maximum possible quantity of the radon adsorbed by this filter during its former use
- a filter initially radon free, used for a time length shorter than its retention time, delivers at the output an almost radon free air (few radon atoms will be present due to the uranium and thorium nuclei making part of the filter).
- using the filter for time lengths shorter than T_r , all the radon molecules temporarily held back by it will never reach the filter output; they will be simply flushed away during the next regeneration cycle.

In order to be really effective, the last solution described above, obviously requires efficient methods to regenerate the filters. Two methods are possible.

The first one consists in cleaning the filter flushing hot air through it, exploiting the strong dependence of the K constant upon temperature (see {3} and {4}).

The second method requires pressurised air to be sent to the decontaminating filter; moreover, a fraction of the clean air produced by this filter will be used to flush the second one. In this configuration the basic mechanism is elucidated by {4} and is linked to the strong dependence of the fraction of radon adsorbed by a filter upon the partial

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pressure of the gas in external air.

Whatever the solution is, the alternate use of two filters requires some energy dissipation, but it implies reduced filters lengths and a higher working temperature. For instance, two filters whose thickness is about *10 cm* could be used at room temperature if they are switched every hour. The same filters, at $0\text{ }^{\circ}\text{C}$, could be cycled every three hours.

Conclusion:

The present study shows that it is possible to realise devices which could be very useful to reduce the environmental radon level of a factor 100 - 1000, in order to fulfill the requirements of low-level background experiments.

Moreover, we would like to underline that the physical processes described here will constitute the basis for the realisation of devices that, concentrating small amounts of radon, can solve the problem of the measurement of low radon level in well defined volumes.