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Radon Diffusion Measurement in Polyethylene based on Alpha Detection

Wolfgang Rau

Department of Physics, Queen's University Kingston, ON, Canada K7L 3N6

Abstract. We present a method to measure the diffusion of Radon in solid materials based on the alpha decay of the radon daughter products. In contrast to usual diffusion measurements which detect the radon that penetrates a thin barrier, we let the radon diffuse into the material and then measure the alpha decays of the radon daughter products in the material. We applied this method to regular and ultra high molecular weight poly ethylene and find diffusion lengths of order of mm as expected. However, the preliminary analysis shows significant differences between two different approaches we have chosen. These differences may be explained by the different experimental conditions.

Keywords: Radon, Diffusion in Polyethylene, Dark Matter

PACS: 66.30.je, 23.60.+e, 25.55.-e, 95.35.+d

INTRODUCTION

Modern experiments in Particle Astrophysics such as direct searches for Dark Matter in the form of Weakly Interacting Massive Particles (WIMPs) which may constitute more than 80 % of the matter in the Universe, searches for neutrinoless double beta decay, which may reveal the absolute mass scale and particle nature of the neutrino, or measurements of low energy neutrinos from different sources are all searching for exotic events with low energies occurring at a very low rate. As such they have very strict requirements regarding the radioactive background radiation which may mimic the rare events searched for.

Radon (in particular ^{222}Rn) poses a serious threat to such experiments due to its high mobility and the long half live of its daughter products. Radon decays through a series of alpha and beta decays into the long lived ^{210}Pb ($t_{1/2} = 22.3$ years), which in turn decays via ^{210}Bi to the alpha emitter ^{210}Po ($t_{1/2} = 138$ days) until it reaches eventually the stable nuclide ^{206}Pb . Besides the direct radiation from the long lived daughter products, there is a chance that the alpha decay of ^{210}Po produces neutrons via (α, n) reactions in the material where it is deposited. Neutrons are of special concern for Dark Matter experiments since the nuclear recoil produced by a neutron is indistinguishable from a nuclear recoil induced by a WIMP.

In this paper we are specifically concerned about the interplay of radon with polyethylene, a material commonly used in dark matter experiments as shield against neutrons. As a first step in determining the potential danger, we investigate the amount of radon that accumulates when polyethylene is exposed to room air. In a later study we will then determine what the neutron production rate from the accumulated radon would be, so we can pose a limit on the allowed exposure of the shielding material to radon.

METHOD

We study the diffusion properties of polyethylene by first exposing the material to a high concentration of radon for an extended period of time. Then we measure the alpha decays of the radon daughter products. Two different incarnations of this method have been tested, one based on the long lived ^{210}Po , and one relying on the short lived products ^{218}Po and ^{214}Po ; in this latter method also the radon itself is observed. This is different than the usually

applied in-situ method of measuring the amount of radon that penetrates a thin barrier [1]. The advantage of our method is that even with a moderately strong radon source it is possible to do the measurement by just increasing the exposure time, while an in-situ measurement requires a fairly strong source. Further, with our first method we directly map out the exponential activity distribution which reduces the sensitivity to systematic uncertainties. On the other hand, the use of ^{210}Po automatically involves long time scales (typically several months before the result can be retrieved).

Long Lived Daughter: ^{210}Po

For this method we prepare a sample of poly ethylene consisting of thin sheets which are carefully cleaned and pressed together into a block of material. One side of this block is then exposed to radon which consequently will diffuse through the different layers. After a certain time (typically a couple of weeks) the sheets are separated and the remaining radon will quickly diffuse out. After an appropriate time during which the ^{210}Pb that was built up while the sample was exposed to radon produces ^{210}Po , the individual poly ethylene sheets are exposed to an alpha detector (ORTEC Dual Alpha Spectrometer 576A) and the ^{210}Po concentration is determined.

Due to its long half life, the ^{210}Po will not be in equilibrium with its parent ^{210}Pb and the relative activity will change over time. Since the individual measurements take of order of days, this change in relative activity has to be taken into account properly. Also, we need to consider that ^{210}Po is started to be produced already during the radon exposure. This is done by a diffusion and decay model which calculates the ^{210}Po activity for any point in space and time for a given radon exposure and given diffusion parameters (we consider the diffusion constant and the solubility, which determines the ratio of the radon concentration on either side of the air-poly ethylene interface). The diffusion parameters are extracted by varying their values in the model until the best fit to the measurements is found.

In addition we determine the efficiency of the alpha detection with a Monte Carlo simulation, based on the simulation package SRIM [2].

Short Lived Daughters

For this method we start the measurement immediately after the end of the exposure of the sample to the high radon concentration. The alpha decays of the short lived daughter products are observed as the radon diffuses out of the material. In this case we only measure one sheet for each sample. We take advantage of the different live times and alpha energies of the involved nuclides by splitting the data into four energy bins. The spectrum changes with the decay of the different species as well as with the change in the depths profile due to the diffusion. We again compare the measurement to a model calculation, but in this case we only extract the diffusion constant. Additional free parameters in the fit are a potential enhancement of radon daughter products on the material surface and the time between end of exposure and the start of measurement (this is particularly critical for the surface contamination with ^{218}Po).

RESULTS

We applied this method to two types of samples: regular poly ethylene (PE) and ultra high molecular weight poly ethylene (HMWPE). In figure 1 you see a comparison between the measurement and the fit for the first method applied to the HMWPE sample. For the purpose of this visual presentation only, the measured values are converted to the equivalent ^{210}Pb concentration at the end of the radon exposure (the fit is done based on the original measurements). The fit quality is reasonable and the extracted parameters are of order of mm as expected for soft polymers [1].

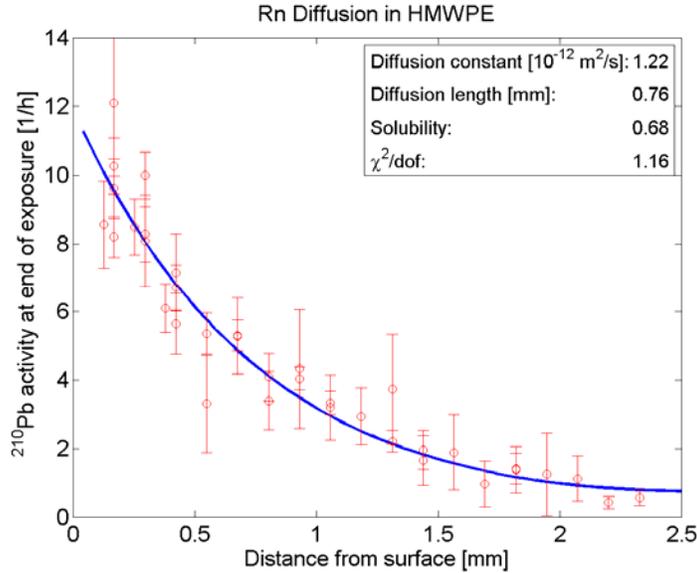


FIGURE 1. Radon diffusion in HMWPE. This measurement is based on the alpha decay of ^{210}Po . To compare the data points with the model calculation (solid line) they have been converted to the equivalent activity of ^{210}Pb at the end of exposure. The fit is reasonable and the extracted diffusion parameters are in the expected range.

In figure 2 you see our model compared to the measurement of the short lived products in HMWPE. The four curves represent the four energy bins of our analysis. In contrast to the first method where the activity is plotted as function of the distance from the surface, here we consider the rate as function of time as the radon diffuses out of the material.

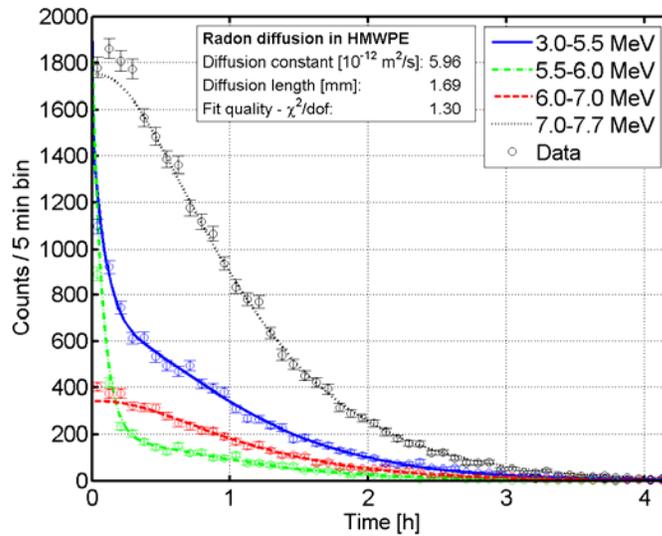


FIGURE 2. Radon diffusion in HMWPE. This measurement is based on the short lived radon decay products ^{218}Po and ^{214}Po as well as on ^{222}Rn itself. To take advantage of the different decay energies and half lives the data have been split in four energy bins as indicated in the legend, before comparing to the rates predicted by the model.

In the table below we summarize our results; the uncertainties are determined by mapping out the χ^2 -surface and finding the values where the reduced χ^2 is one unit above the value of the best fit. Note that the uncertainties in solubility and diffusion constant are correlated and the resulting uncertainty in the total amount of radon accumulated for a given exposure is smaller than a naïve combination of the uncertainties would imply.

Please note further that all results and uncertainties presented in this paper are preliminary and require further refinement, however neither the central values nor the uncertainties are expected to change significantly.

TABLE. Diffusion parameters for PE and HMWPE as measured with the two different methods described in this paper. For a discussion of possible reasons for the differences between the results of the two methods see text.

Sample	²¹⁰ Po			Short lived	
	Solubility	Diff. Const. [10 ⁻¹² m ² /s]	Diff. length [mm]	Diff. Const. [10 ⁻¹² m ² /s]	Diff. length [mm]
PE	0.68 ± 0.10	1.2 ± 0.2	0.76 ± 0.06	6.0 ± 0.8	1.7 ± 0.1
HMWPE	0.45 ± 0.07	3.7+1.4/-1.0	1.3 ± 0.20	7.1 ± 0.8	1.8 ± 0.1

We notice that the diffusion constants found by the two different methods do not agree. There are a number of possible reasons for this observation: it is known that humidity can greatly influence the diffusion of radon in polymers [1]. The humidity of the samples was not controlled in this measurement, which may lead to differences in the results from the two methods. Another possible contribution to the differences may be the fact that the samples are under vacuum while exposed to the alpha detector. For the first method (²¹⁰Po measurement) the radon diffuses into the sample under normal pressure, while for the second method (short lived decay products) the radon diffuses out of the material under vacuum. The low pressure may pull other gases out of the sample which in turn could enhance the diffusion of the radon atoms. With the available data no further conclusion about the origin of the differences can be drawn.

CONCLUSION

In this paper we have presented two methods for the measurement of radon diffusion in polymers based on the detection of alpha decays from radon daughter products. The diffusion parameters are extracted based on the comparison of the measured data with a diffusion and decay model and a Monte Carlo simulation of the efficiency of the detection of the alphas from the different daughter products. We find a reasonable agreement between the behavior predicted by the model and the measurements; however, we find significant differences in the extracted values between the two methods, which may be explained by the different experimental conditions.

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