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A novel detection of radon based on its decay product inducing conformational changes of an aptamer probe

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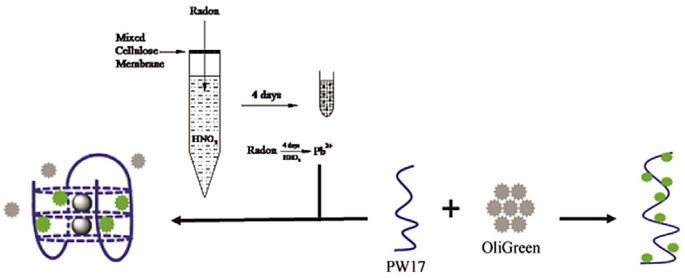
HIGHLIGHTS

- The label-free fluorescence sensor for detection of radon.
- This microscale experiment without radiation damage to experimenters and with less harm to environment.
- It provides a sensitive, low cost and simple strategy for radon accumulated concentration and lead ion detection.

GRAPHICAL ABSTRACT



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ABSTRACT

This study proposes a novel method for the detection of inert gas radon using a label-free, specific, fluorescence-sensing aptamer in the context of PW17-OG system. This method utilizes the cyanine dye OliGreen (OG) as a signal reactor and the aptamer PW17 as a fluorescent identification probe. When OG integrates into the free curling PW17, a strong fluorescence signal is generated. After radon decays, the long lived naturally occurring radon progeny Pb being disposed and introduced to the system. Lead ions induce PW17 to form a stable G-quadruplex, thereby inhibiting the interaction between OG and PW17 and resulting in a reduction of the fluorescence intensity. The fluorescence intensity show a good linear relationship with lead ion and the radon concentration (D), thereinto, We fitted linear regression of radon concentration in the range of 0.92–4.22 ($\times 10^4$ Bqhm^{-3}) to receive a good relationship between ΔF and the concentration of radon with the detection limit of 1963 Bqhm^{-3}. This method has been successfully applied for detecting standard cumulative concentration of radon and the detection limit reached the national standard of China. This sensitive method can exclude radiation damage in field testing, furthermore, it explores a new field in biological analysis using an aptamer to detected inorganic, gaseous, and radioactive materials.

1. Introduction

The radioactive noble gas ^{222}radon is produced by the decay of ^{226}radium within the natural decay chain of ^{238}uranium [1]. The following ^{222}Rn (half-life 3.82 d) decay chain is ^{218}Po (3.05 min), ^{214}Pb (26.8 min), ^{214}Bi (19.7 min), ^{214}Po (1.64×10^{-4} s), and ^{210}Pb (21 y) [2]. Radon is commonly found in all rocks and soil [3] and the byproducts can cause substantial damage to the respiratory, hematopoietic, and digestive systems; in severe cases, it may lead to lung cancer [4]. Humans have constant exposure to radon, therefore, the sensitive and reliable detection of radon concentrations is of particular interest to public health.

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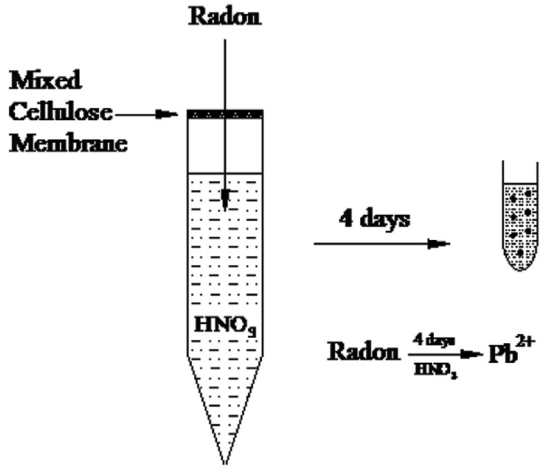
Currently traditional methods for detecting α and β-rays generated from radon radiation [5] include activated charcoal absorption, α-track etch detection, and electret probing [6,7]. However, these strategies are limited by various factors including the unwieldy size and exorbitant price of the required equipment, a relatively protracted period of detection, and health risks to personnel carrying out the experiments. Radon is persistent continues to damage to the human body, as a result, an urgent need exists for a new method that simplifies the operation of detecting the accumulative doses of radon. This method have solved those problem above, and may have the potential for application. The sampling device with small size is easy to transport, and it can be mailed to the tested area directly after subsequent improvements.

Over the past decades, the discovery of aptamers has shown tremendous potential and versatility in the analytical and bioanalytical fields [8]. By focusing on the decay product generated from radon radiation and these findings, our group pioneered a new approach to detecting radon using biological analysis. The potential contamination was prevented from entering the tube by covering mixed cellulose membrane with 0.8 μm pore which can block 99.9% of the already exists radon progeny in the laboratory air, allowing only radioactive particles and gas molecules into the samples [9,10], and achieving highly purified samples as a result.

We were putting the sampling equipment in radon chamber; the equipment were exposed under the standard concentration of radon. During the sampling of radon, standard concentration of radon in radon chamber diffused into the sampling equipment. By controlling the exposure time to getting the corresponding standard cumulative concentration of radon. After radon collects, the samples would have gone through a half-life cycle to further stabilizing the sample system and gathering more considerable stable daughter Pb to oxidizing to Pb^{2+}(Scheme 1).

Meanwhile, according to the previous reports [11–13], We selected the oligonucleotide PW17, which was enriched with guanine and used as an identification probe, with OG acting as the signal reactor. The cyanine dye OliGreen (OG) is scarcely fluorescent because its quantum yield is less than one percent [14]; however, when it binds to single-strand oligonucleotide, its fluorescence is amplified 1000 times [15]. Although OG interacts strongly with free single-strand aptamers, its fluorescence intensity changes as the aptamer structure transforms [14–16]. We presumed that the interactions between OG and PW17 produce a strong fluorescence (Scheme 2). When radon samples or Pb^{2+} is introduced to the reaction, it causes the formation of a stable G-quadruplex structure from unbound state, thereby inhibiting the interaction between PW17 and OG. The loss of this interaction results in a significant reduction in fluorescence intensity. Based on non-labeled fluorescent intensity, this method successfully achieves sensitive detection of lead and radon in the environment. The present study explores a new field in biological analysis by using aptamers to detect non-metallic radioactive gases.



Scheme 1. The principle diagram of the sample processing.

2. Experimental

2.1. Apparatus

This study was conducted in a radon chamber at South China University's Institute of Nuclear Industry No. 6. The experiments utilized an F-4500 fluorescence spectrophotometer (Hitachi, Japan), with 1200 nm/min scanning speed, 5 nm and 10 nm slits for excitation and emission wavelength respectively. We also used a circular dichroism JASCO-815 instrument (JASCO Corporation, Japan), with a scanning speed of 200 nm/min.

2.2. Reagents

PAGE-purified oligonucleotide: PW17 (5′-GGGTAGGGCG GGTTGGG-3′); R (5′-ATCGAAATTCGCATCGG-3′) was purchased from Sangon Biotech Ltd. (Shanghai, China), and diluted with ultrapure water to a concentration of 100 μM and stored at 4 °C until needed. The OG dye used (Molecular Robes Corporation) has a defined concentration of 2000×, and it was stored at −20 °C.

The standard lead solution used (China Institute of Metrology) was diluted with ultrapure water to a concentration of 100 μg/L of Pb^{2+} in the working solution. Trisaminomethane (Tris) with a purity of at least 99.9%, purchased from Shanghai Aladdin Biochemical Technology Co., Ltd., was used to form a Tris-HNO_3 buffer as the same with radon samples and pH of 7.0. Premium grade nitric acid was purchased from Sinopharm Chemical Reagent Ltd. Co. $Bi(NO_3)_3 \cdot 5H_2O$, acquired from Guangdong Guanghua Sci-Tech Co., Ltd., had a purity of at least 99%. All reagents were diluted with ultrapure water and stored at 4 °C.

2.3. Procedure

2.3.1. Preparing of samples

We added 14 mL of 0.2%HNO_3 to a series of 15 mL centrifugation tubes, which were covered with a mixed cellulose membrane (Merck Millipore Ltd.) with an aperture of 0.8 μm and fixed with adhesive tape. The tubes were then exposed to radon laboratory for sampling. By controlling the exposure time to getting the corresponding standard cumulative concentration of radon. After sample retrieval, the tubes were sealed at room temperature for four days before measurement. Four days later, the sample mixed with Tris buffer to reach a pH level of 7.0 before measurement.

2.3.2. Measurement of Pb^{2+} concentration

Pb^{2+} working solution was added to a series of centrifugation tubes to yield a gradient solution of final concentration from 0 to 35 μg/L with 20 μL of 1 μM PW17, 75 μL of Tris-HNO_3, and filled each tube with distilled water to 180 μl. The mixture were incubated at room temperature for 30 min before the addition of 20 μl of 20 × OG dye. Fluorescence intensity was measured at an excitation wavelength of 500 nm and an emission wavelength of 524 nm. A standard curve using different Pb^{2+} concentrations and a linear regression equation was calculated based on the findings for the fluorescence intensity.