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The Atmospheric Chemistry of Po-218

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Abstract

The chemical and physical properties of ^{218}Po immediately following its formation from ^{222}Rn decay are important in determining its behavior in indoor atmospheres and plays a major part in determining its potential health effects. In 88% of the decays, a singly charged, positive ion of ^{218}Po is obtained at the end of its recoil path. The neutralization rates for Po-218 by the small ion recombination, electron transfer or electron scavenging mechanisms were previously reported. We have measured the small ion recombination rate in high purity gases to determine the negative small ion production rate as a function of the ionization potential of the gas. Our previous studies have also shown that radiolysis of water vapor by the recoiling Po ion produces high local concentrations of hydroxyl ($\cdot\text{OH}$) radicals. These radicals can lead to oxidation of reactive trace gases and the neutral polonium molecule can become incorporated in ultrafine particles formed by the nucleation of low vapor pressure compounds produced by this radical oxidation process. The hydroxyl radical production rate and the production of particles are currently being studied.

INTRODUCTION

The health effects of the radon decay products are strongly dependent upon their behavior in the indoor atmosphere. Particularly for ^{218}Po , the partitioning between the "unattached" state and the "attached" forms, combined with the preexisting aerosol particles, has a significant impact on the calculation of the dose to the lung resulting from a given decay product concentration. Thus, the chemical and physical behavior of the ^{218}Po atom immediately following its formation by the alpha decay of radon must be considered if its health implications are to be fully assessed.

NEUTRALIZATION RATE MEASUREMENTS

At the time when the alpha leaves the nucleus, there are two excess electrons around the residual polonium nucleus. It is probable that the exiting alpha causes autoionization leading to a positive ion. The recoiling polonium nucleus has a recoil energy of 117 keV with a range in air of approximately 70 microns (Lind, 1961). The ion will be gas stripped to a moderate positive charge and then regain electrons as it slows toward thermal velocity such that at the end of the recoil path, 88% of the polonium atoms have a +1 charge and the remaining 12% are neutral (Wellisch, 1913; Porstendorfer and Mercer, 1979). The study of the neutralization behavior of the polonium ion has been a major focus of the initial efforts of this project. The results of these studies have been reported by Chu and Hopke (1988). Their results can be applied to the understanding of the electrostatic collection of radon decay products particularly in radon measurement systems based on such collection. These studies have been described by Hopke (1989a). Thus, we now have a quantitative understanding of the neutralization behavior of $^{218}\text{Po}^+$ in indoor air. To complete the basic studies of $^{218}\text{Po}^+$ neutralization, we have also measured the rate of neutralization by small ion recombination in a series of ultrapure gas atmospheres including oxygen, nitrogen, and argon (Shi, 1989).

RADIOLYTIC NUCLEI FORMATION

As indicated previously (Ramamurthi and Hopke, 1989), the health effects of the radon decay products are strongly dependent upon the partitioning of the radon progeny between the "unattached" state and the "attached" forms. The differences in the diffusivity of these activity size modes has a significant impact on the calculated dose to the lung from a given decay product concentration. Previous studies had focussed on the measurement of an

operationally defined "unattached" fraction. Ramamurthi and Hopke (1989) have reviewed the prior use of wire screens for the separation of the "unattached" fraction. They found that the choice of a single, optimized screen $d_p(50\%)$ parameter that maximizes the collection of the "unattached" mode while minimizing the collection of "attached" activity may be beneficial for single screen "unattached" fraction measurements. However, it is even better to measure the entire activity size distribution and then determine the activity fraction within the highly diffusive region of the size spectrum. The development of such a system is the focus of our other DOE/OHER project (Ramamurthi and Hopke, 1990). Our prior studies on radiolytic nuclei production have been summarized by Hopke (1989b). During the past year, we have pursued both experimental and theoretical studies of particle formation in our 2.4 m³ radon-aerosol chamber (Karpen-Hayes and Hopke, 1990; Lao *et al.* 1990).

OXIDATIVE RADICAL PRODUCTION

A key factor in determining the role of radon radiolysis in indoor atmospheric chemistry is the concentration of $\cdot\text{OH}$ radical averaged over the entire air volume. Chu and Hopke (1988) estimated the $\cdot\text{OH}$ concentration in the immediate vicinity of the recoiling polonium atom but made no estimation for the average bulk concentration. Both the α and the recoil paths would contribute to this $\cdot\text{OH}$ concentration. Studies are in progress to examine the $\cdot\text{OH}$ concentration using the chemical multiplication chemiluminescence method of Cantrell *et al.* (1984). We have found a linear response of the chemiluminescent signal to changes in radon concentration in a mixture containing 10% CO, 10 ppm NO, 20% O₂, 40% relative humidity ($\sim 10,000$ ppm H₂O), and the remainder being N₂ (Figure 1). We are working to calibrate this response in terms of the $\cdot\text{OH}$ concentration as well as measuring the response to variable H₂O concentration at fixed radon levels. These studies will lead to an understanding of the rate of hydroxyl radical production that is occurring in indoor air as a result of radon radiolysis.

SUMMARY

The project has succeeded in achieving its initial objectives. We now have quantitative measurements of the rates of neutralization of $^{218}\text{Po}^+$ for each of the various mechanisms that are available. These measurements have permitted the calculation of the effects of neutralization of the collection efficiency of the radon progeny, particularly in radon

Figure 1. Relative Response of the Fluorescence Detector to the NO₂ Chemiluminescence.

detectors that are based on the electrostatic collection of the decay products (Hopke, 1989a). We have improved our understanding of the formation of radiolytic nuclei and have shown that we can obtain a measurable response from the system design to determine the free radical production rates. We are now in a position to begin to obtain quantitative information of production rates of both free radicals and ion-induced nuclei. These studies will be continued under the new Department of Energy grant (DE FG02 90ER61029) that began July 1, 1990.

PROJECT PERSONNEL

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PUBLICATIONS ARISING FROM THIS CONTRACT

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PRESENTATIONS OF RESULTS OF THIS CONTRACT

The Measurement of Size Fractions of Radon Progeny, P.K. Hopke, presented at the Technical Measurements Center, operated by UNC Geotech for the Grand Junction Project Office, Grand Junction, Colorado, September 1989.

Formation of Ultrafine Aerosols (0.5-20 nm) from Radon Radiolysis. P.K. Hopke and M. Ramamurthi, R. Strydom, K.W. Tu, E.O. Knutson, R.F. Holub, W. Winklmayr, W. Marlow and

S.C. Yoon, presented at the American Association for Aerosol Research, Reno, NV. October 1989.

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Potential for Forming Ultrafine Aerosols from Indoor Organic Vapors by Radon Decay Radiolysis-Induced Reactions, J.M. Daisey and P.K. Hopke, presented at AAAR 1990 Annual Meeting, PA., June, 1990.

Production of Sulfuric Acid Aerosol by Radiolytic Conversion of SO₂, K. Karpen-Hayes and P.K. Hopke, presented at AAAR 1990 Annual Meeting, PA., June, 1990.

Measurement of the Effect of SO₂ on the Polonium-218 Ion Mobility Spectrum by Alpha-Track Detection, S.C. Yoon, W.H. Marlow, P.K. Hopke, and M. Ramamurthi, presented at AAAR 1990 Annual Meeting, PA., June, 1990.

Modeling Radon Progeny Size Distribution in An Aerosol Chamber, M. Lao, K. Karpen-Hayes and P.K. Hopke, presented at the American Chemical Society Northeast Regional Meeting, June, 1990.

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