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DESIGN OF A RECIRCULATING RADON PROGENY AEROSOL GENERATION AND ANIMAL EXPOSURE SYSTEM

PRINCIPAL INVESTIGATORS

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Abstract — *Inhalation studies are being conducted at ITRI using laboratory animals exposed to radon-222 progeny attached to vector aerosols that are typical of indoor environments. The purpose of these studies is to identify the cells at risk from inhaled radon progeny and their locations within the respiratory tract. These studies require exposures up to 1000 working level months (WLM) within a few hours. Thus, large amounts of radium-226 are needed to produce the gaseous radon-222. A once-through-exposure-system was considered to be impractical because of statutory discharge limitations and the large amounts of radium that would be required. Therefore, a recirculating exposure system was designed and constructed that removes the aerosol after passing through the exposure chambers and recirculates purified air and radon. The purified radon is mixed with freshly evolving radon from a radon generator and passed into a reaction-aging chamber where attachment of radon progeny to the vector aerosol occurs. The design includes: (1) 50-200 mg radium-226 in a radon generator, (2) 40 L/min total flow rate, (3) CO_2 removal, (4) reconstitution of oxygen tension and water vapor content to atmospheric levels, and (5) a trap for radon gas. A radon progeny exposure concentration in the range of 4,000 to 50,000 WL is being produced.*

To identify the lung cells at risk from exposure to radon progeny aerosols and their locations within the respiratory tract requires a laboratory animal exposure system capable of delivering an exposure up to 1000 WLM in about 5 h. The required chamber operating concentrations are about 40,000 WL. This required construction of a unique aerosol generation and exposure system that is safe and practical to operate.

Simplified approaches for radon progeny aerosol generation for inhalation exposures of laboratory animals are shown in Figure 1. The systems use a dry radium source that releases radon gas into an aging chamber upon ventilation. Because radon-222 decays with a 3.83 day half-life, and its biologically significant progeny decay with half-lives ranging from 3.05 min to 26.8 min, an aging chamber is required to hold the radon and progeny until an acceptable level of equilibrium is obtained. For an equilibrium factor, F, of 0.5, the holdup time required for ingrowth of the radon progeny is about 30 times the minute volumetric flow rate through the system. These approaches for an exposure system have two main options: (1) a once-through aerosol system followed by radon trapping (Fig. 1A) or (2) a closed loop system to recirculate radon (Fig. 1B). The maximum air concentration of radon permitted for discharge to the atmosphere is 1.0×10^{-9} µCi/mL. Because our inhalation exposure system requires about 6.8 µCi of radon-222 per liter, this eliminated the option of a once-through-system without a radon trap because the dilution volumes could not be attained without unrealistically high flow rates. A radon trap for a once-through-system was also considered impractical because of its size and shielding requirements.

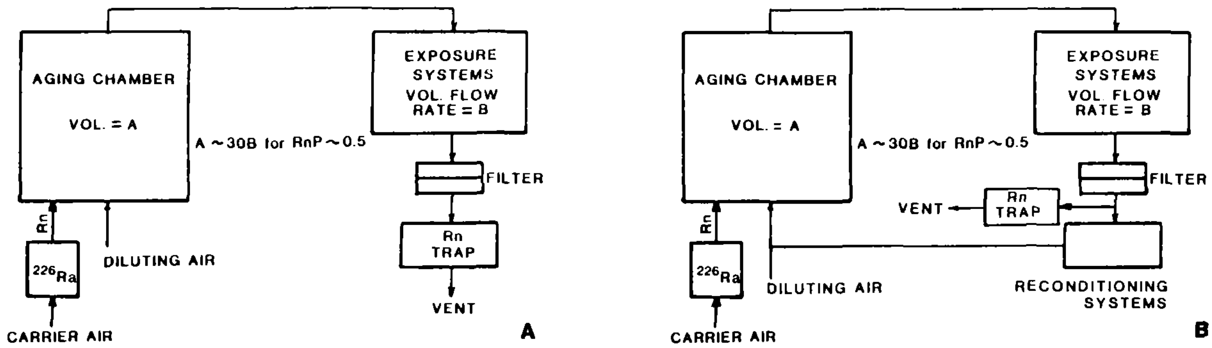

Figure 1. Schematic diagram of a simplified radon radon-progeny aerosol generation and animal inhalation exposure system. Figure 1A shows a once-through-system and Figure 1B shows a closed loop or recirculating system.

To meet the programmatic goals, satisfy the radon discharge limits, and operate the system with a minimum amount of radium, we examined an aerosol exposure system based on principals discussed by Morken.[1] To achieve required levels for exposures of rodents to pure radon-222, Morken developed a closed loop system. Exhaust from the exposure system was: (1) filtered to remove the particulate species, (2) reduced in humidity by adsorption of water vapor, (3) passed over a carbon dioxide adsorber, (4) monitored for oxygen content, and (5) oxygen content restored to desired levels by addition of pure oxygen.

Conceptual Design of the Recirculating Radon Progeny Aerosol System

For the above reasons and also because of limited availability of ^{226}Ra, a recirculating system was designed (Fig. 2). Although more complex, a recirculating radon system resulted in reduced radium requirements and reduced radon trapping requirements with attendant savings in shielding and other safety concerns.

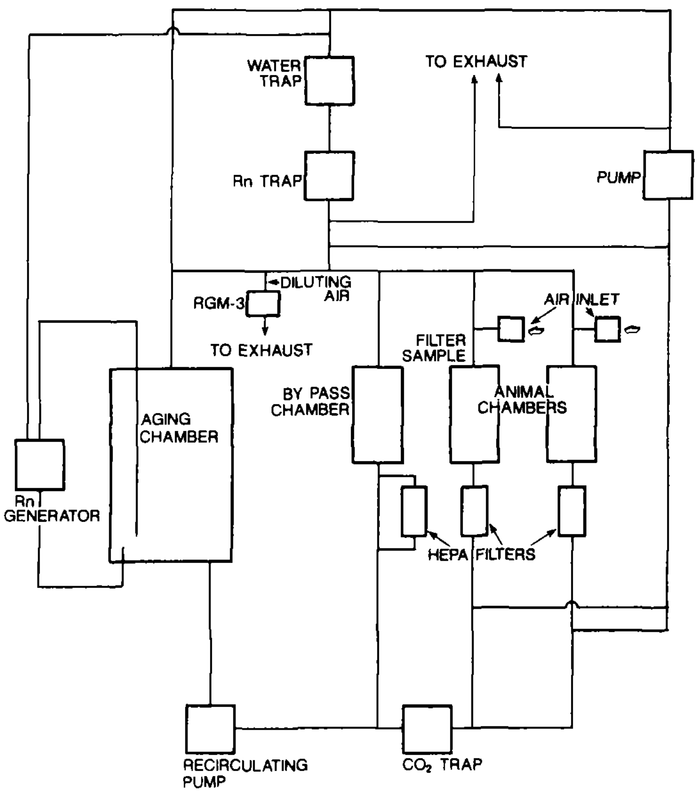

Figure 2. Schematic diagram of the recirculating closed-loop radon progeny aerosol generation and animal inhalation exposure system.

Essential elements of the small animal inhalation exposure system are: (1) a shielded filter assembly to remove entrained particles and the associated radon progeny, (2) a water removal system, (3) a CO_2 trap, (4) a radon trap, (5) an oxygen monitor, and (6) an oxygen injection system. The oxygen injection system restores the exhaust to acceptable breathing air standards. The reconstituted air (with most of the radon still present) is compressed to about 20 psig. The compressed air-radon mixture is used to aspirate the vector aerosol generator. This re-enters the aerosol-aging chamber and the exposure atmosphere. Gases to be vented from any part of the system pass through a radon trap and are held for decay, thus minimizing the environmental release of radon-222.

The radon-222 aerosol generator consists of an insoluble radium stearate[2] source in a porous stainless steel tube which is inside a brass pipe (Fig. 3). The radon generator is shielded with lead bricks in a glove-box. An airstream passes through the porous metal tube entraining gaseous radon. Radon-222 in the airflow enters the 2 m^3 aerosol-aging chamber.

Vector aerosol particles are mixed with the radon and radon progeny in the aging chamber. The small radon progeny attach to the vector particles according to the dynamics of the attachment process. The aerosol-aging chamber provides 30-50 min of residence time to increase the concentrations of the radon progeny. From the aerosol-aging chamber, aerosol is piped to the animal inhalation chambers.

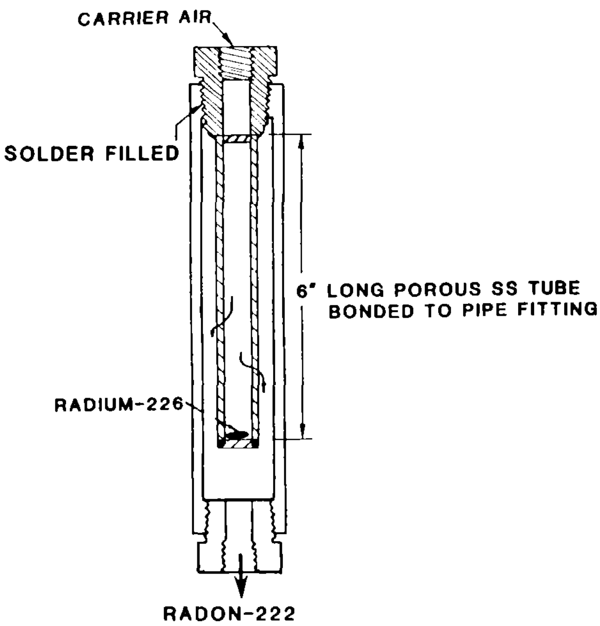

Figure 3. Sketch of the container used to hold radium and release gaseous radon.

The volume of the system consists of a 2 m^3 aging chamber, a 1 m^3 bypass-sampling chamber, and a 1 m^3 animal exposure chamber. The pre-exposure setup involves cycling aerosols through the aging chamber and the bypass-sampling chamber. When a stable exposure aerosol is obtained, the bypass-sampling chamber is isolated and the radon-radon progeny-containing aerosol is passed through the animal exposure chamber. At shutdown of the animal inhalation exposure, the bypass-sampling chamber is put back into the recirculating loop and the animal exposure chamber is isolated from the radon sources. The purged atmosphere that contains about a quarter of the radon in the entire system is exhausted through the radon trap. After trapping the radon, the animal exposure chamber is purged with clean air and the animals are removed.

The radon trap consists of two parallel charcoal canisters operated at 2°C.[3] After water removal, the purge airstream is passed through a charcoal bed that adsorbs radon. The radon trap containing the adsorbed radon can be heated to release the trapped radon to be reused if required.

Because the aerosol generation system contains several components that could collect significant quantities of radon progeny that emit energetic gamma photons, most of the system was placed behind an 8-inch thick solid concrete block wall. The main components of concern were exhaust filters from the animal exposure chamber, the exposure chamber, the aerosol aging chamber, the radon trap, and the air compressors. Figure 4 is a sketch of the floor plan showing the layout of the major components.

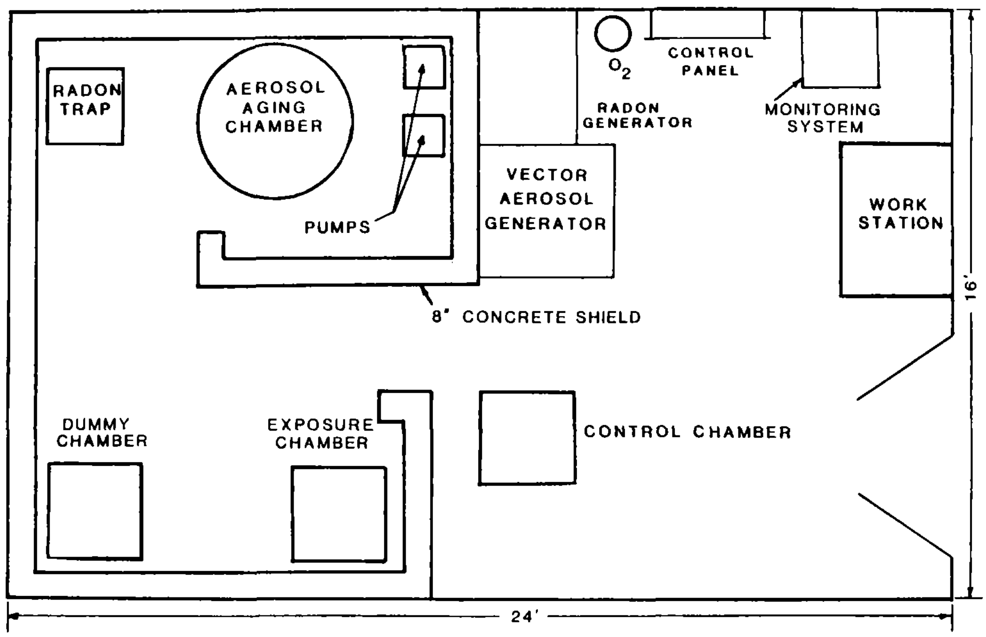

Figure 4. Floor plan of the radon progeny exposure laboratory showing location of the major components of the systems.

VECTOR AEROSOLS

Vector or carrier aerosols that will be used for attachment of radon progeny will include types of particles, size distributions, and airborne mass concentrations relevant to human radon exposure situations. Of prime importance are aerosols typical of early uranium mines. Generally, these are the older and smaller mines of the Colorado plateau that had high dust loadings. Many of the mines relied strongly on muscle power although by the 1960s some of the mines used underground diesel powered equipment and others used electrical systems. For the mines with underground diesel powered equipment, the aerosol size distribution would have been multi-component, multi-modal with a small size mode of less than about 0.1 µm diameter and larger mode(s) in the 1-10 µm aerodynamic diameter range. We are simulating these multi-modal aerosols with mixtures of particles in the range of 1-10 µm mass median aerodynamic diameter (MMAD), generated by systems that include dry powder dispersion of uranium ore dust or nebulization of a uranium ore dust simulant. The ultrafine particle size mode (< 0.1 µm) is either diesel particles or cigarette smoke or both as a mixture. Other relevant vector aerosols associated with indoor air are cigarette smoke and fumes from cooking.

AEROSOL CHARACTERIZATION

Aerosol characteristics for estimating deposition and dose to the cells at risk will be determined. These include: (1) the working level, WL, (2) radioactivity size distributions of radon progeny, (3) number, size, and mass concentration of the inert vector particle size distribution, (4) relative equilibrium factor, (5) concentration of unattached radon progeny, and (6) concentration of radon gas. To obtain these aerosol parameters, a battery of aerosol instruments will be used. The combination sizing instrument that consists of the Lovelace Multijet cascade impactor (LMJ) and the parallel flow diffusion battery (PFDB) provides fourteen size cuts from about 10 μm aerodynamic diameter down to about 0.05 μm diffusion diameter. Seven size cuts from 10 μm to 0.7 μm aerodynamic diameter can be obtained from the LMJ. The PFDB can provide equivalent diffusion diameters in the size range from 0.7 μm to about 0.05 μm (six size cuts plus a final filter). Aerosol parameters for the vector particles will be determined from the LMJ/PFDB samples along with complimentary filter samples and number concentration data from commercially available continuous flow condensation nucleus counters. Other instruments will provide data on the WL, radon and radon progeny concentrations. The equilibrium factor, F, will be determined from analysis of two specially designed filter samplers. One filter sample is taken directly onto the filter material, the other parallel sample is preceded by a single 200 mesh screen that is about 99% efficient for collection of unattached radon progeny. Radon progeny attached to vector particles penetrate the screen with an efficiency of 99.5%. From the differences in collected radioactivity, we can estimate the unattached radon progeny fraction. Figure 5 is a sketch of these unattached radon progeny samplers.

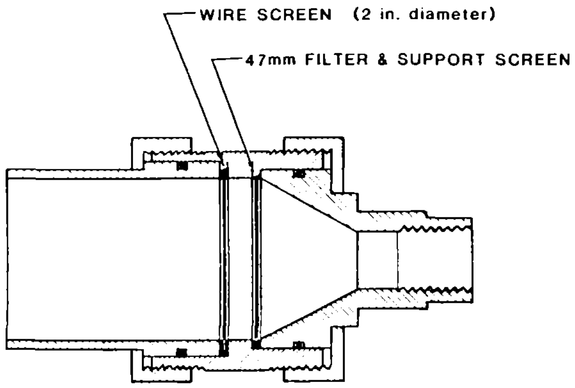

Figure 5. Sketch of the aerosol samplers designed to obtain filter samples for determination of the unattached fraction of radon progeny aerosols. One sample is obtained without using a screen and the other sample is taken simultaneously through the 200 mesh screen.

CURRENT STATUS

The aerosol generation and small animal inhalation exposure system described has been engineered, constructed, leak tested, and used to expose rats to 4,000 WL. No leaks were determined in any of the pressurized components or in any of the piping systems between the exposure chambers. Operating pressures (inches of waters) associated with the various components are: aging chamber = 2 inches, animal exposure chamber = - 6.9 inches, and bypass-sampling chambers = - 4.2 inches. The system has been undergoing operation with a 10 mg radium-226 source to gain operational experience and determine important operational factors that require empirical calibration. Upon completion of the low level testing, a 50 mg source of radium-226 will be used to obtain concentrations up to 40,000 WLs.

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