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THE REDUCTION OF AIRBORNE RADON DAUGHTER CONCENTRATION BY PLATEOUT ON AN AIR MIXING FAN

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Abstract—A series of experiments have been made in the U.S. Bureau of Mines Radon Test Chamber to study the effects of condensation nuclei, humidity and turbulence on the rapid deposition or plateout of radon daughter activity on the chamber walls. Under low humidity conditions the presence of a small fan reduced the working level by 41%. The activity was not deposited on the walls by the turbulent flow from the fan but actually became attached to the fan blades. High relative humidity (>80%) totally inhibited this observed effect. A detailed mechanism for transport of the daughter species seems to be the critical factor in interpreting the experimental results.

INTRODUCTION

It has been known for some time that there is a tendency for the radon daughter activities to attach rapidly or plateout on the surfaces (walls, ceiling, machinery, etc.) of uranium mines (Ho57). A review of the literature by Cooper *et al.* (Co73) indicates a considerable variation in the reported effects of charge, humidity and the number of condensation nuclei on the airborne radon daugher concentrations.

Postendorfer (Po68) measured the diffusion coefficient for charged and neutral ^{212}Pb and found a 33% reduction on the diffusion coefficient of the charged atom relative to the neutral species. These constants were determined 1–5 sec after the formation of the ^{212}Pb atom. The reduction in diffusivity is proposed to be the result of charge effects increasing the cross section for cluster formation between atoms and ions. These species, being heavier, will diffuse more slowly. There was no consideration of the effects of humidity on the measurement.

Raabe (Ra68) has reported results of humidity effects on the diffusion coefficient for RaA in which the diffusion coefficient is reduced from 0.047 cm^2/sec for a dew point of −4°C to 0.034 cm^2/sec for a dew point of 9°C. The experiment examined the effects of a high electric potential to remove ionic species and found no effect; hence, it was concluded that neutral species were being studied. The RaA was sampled from a large air volume having a high randon concentration (1.5×10^{-8} Ci/l.). The air ionization which results from the high level of radioactivity would be expected to cause neutralization which may not occur at much lower radon concentrations.

In contrast, a number of groups (Ch56; Me69; Ko76) have found little effect of humidity on the diffusion coefficient. However, there is general agreement (Ko76; Co56; Me69) that there is a strong dependence of plateout upon the fraction of activity that is attached to particles.

In order to study the effect of turbulent air flow on plateout in mine environments, Wrenn *et al.* (Wr69) and Shreve and Cleveland (Sh72) studied the effect of placing an air-mixing fan in the mine and observing its effect on the working level of daughter activity. Both investigations assumed that their observed reduction in airborne activity was the result of increased plateout on the mine surfaces.

In order to study surface plateout in detail, a radon chamber has been constructed that can be used to test the effects of condensation nuclei, humidity and turbulence on the rate of plateout. This paper reports some of the initial results of studies conducted utilizing this system.

EXPERIMENTAL SETUP

The U.S. Bureau of Mines radon environment chamber was constructed to permit studies of radon and daughter activities in a controlled environment and to test and calibrate instrumentation for monitoring mine atmospheres. The system is shown schematically in Fig. 1. The polydispersed nichrome aerosol is humidified and then mixed with radon from a dry source. The dry source was prepared by coprecipitating Ra ions from a $RaCl_2$ solution with a barium stearate. The emanation coefficient is up to 90%. It is dispersed into the approx. 1800-l. chamber through a planar baffle with 1.0 cm holes spaced at 5.0 cm intervals. In the interior, instrumentation is available to measure the working level with a continuous working level detector (D2), the activity on the walls with a GM-type gamma ray detector (D1), the airborne radon concentration with a continuous scintillation detector (RN), the number of condensation nuclei with an "Environment-1" condensation nuclei counter, and the relative humidity (RH). The air is vented through an exit baffle identical to the inlet system. In the experiments to be discussed, and "IMC Boxer" fan (approx. 3500 rpm) was placed in the lower corner of the chamber. A GM detector (D3) was used to monitor the gamma ray activity on the fan. Thus, all of the activities are measured directly and continuously. The time response of this system to changes in chamber conditions is limited by the nature of the detectors. The continuous working level detectors have been described by Droullard and Holub (Dr77).

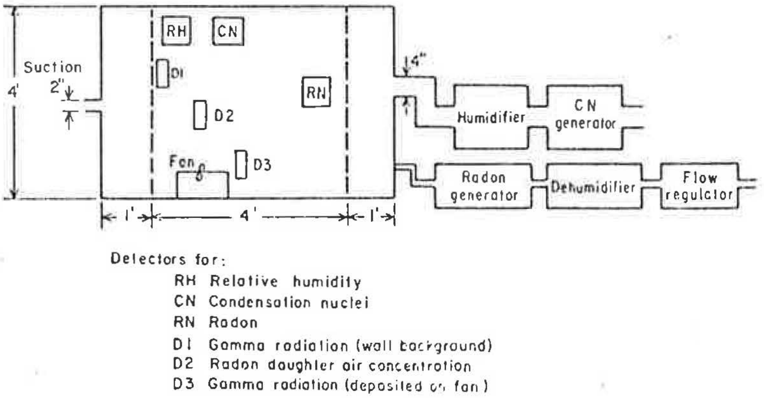

FIG. 1. Schematic diagram of the radon chamber.

RESULTS

A series of anemometer readings were taken to ascertain the pattern of air movement caused by the fan. These results are shown in Fig. 2. With the air flow through the chamber at 72.5 lpm, the fan would recirculate the air within the chamber up to 20 times before it exits the enclosure.

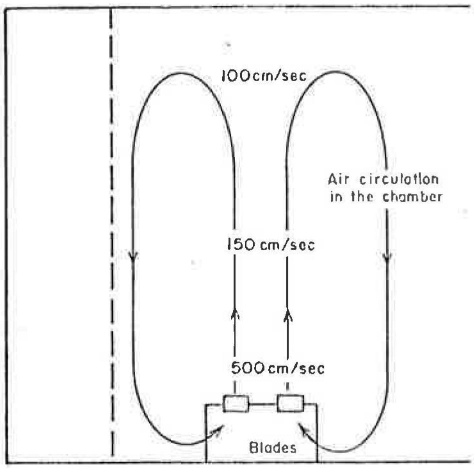

FIG. 2. Sectional view of the fan in the chamber with the air velocity profile.

In order to observe the effect of condensation nuclei on wall deposition, an experiment whose results are shown in Fig. 3 was performed. The system was run with a constant radon level (curve 4) but without the fan or the aerosol generator. After 17 hr, the aerosol generator was started, resulting in a rapid rise in the number of condensation nuclei (curve 3). It can be seen that as the condensation nuclei level decreased during the evening (primarily becuase of the lack of human activity in the area of the system), the working level (curve 1) decreased and the wall background (curve 2) rose. Thus, increased plateout was occurring when the condensation nuclei level became sufficiently low to maintain a large unattached daughter fraction. Once the condensation nuclei count rose to a relatively high level ($10^5/cm^3$), the working level rose and the wall background decayed to lower values, indicating attachment of the daughter activity to the particles, thus preventing wall plateout. These results agree with earlier results (Sh72) in that the absence of particles increases the rate of plateout.

The second experiment was to observe if

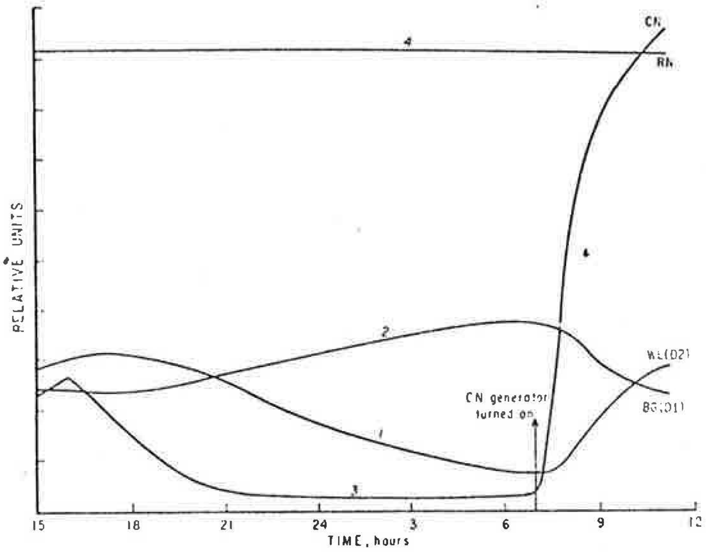

FIG. 3. Radon daughter concentration in the air (curve 1, full scale 10 WL/l.) and on the walls (curve 2, full scale 1000 counts/min) in response to change in condensation nuclei concentration (curve 3, full scale $10^5/cm^3$). Curve 4 (full scale 2500 pCi/l.) is the radon concentration in the air.

the effects of additional turbulent mixing by the fan would affect the rate of wall deposition in the presence of a high condensation nuclei concentration and low relative humidity, to observe if the results of Wrenn *et al.* (Wr69) could be extended to higher particle concentrations. The results of this experiment are shown in Fig. 4. Starting the fan lowered the measured working level (curve 1) by a factor of 2. However, there is no corresponding increase in activity on the wall (curve 2). The activity level that increases is that on the fan itself (curve 3). The action of the fan is to remove airborne radon daughter activity by deposition onto the blades of the fan. It should be noted that although there are variations in the level of condensation nuclei (curve 4), there is no correlation with the presence or absence of fan motion. The radon concentration is unaffected (curve 5). When the fan is stopped, the working level activity increases and the fan activity decreases. This experiment was repeated 6 months after the initial trial with identical results. Wrenn *et al.* (Wr69) apparently did not examine their fan for residual activity following their experiment.

Final experiments were made with the identical conditions except that the humidity was raised to >80%. Under these conditions the working level and fan activity are unaffected by the fan motion. The presence of the water vapor apparently inhibited the attachment of the activity to the moving fan blades. Thus, these experiments indicate that turbulent mixing can result in increased plateout, although the plateout occurs on the fan and not on the walls; that the presence of moisture plays an important role in permitting the activity to become attached at low relative humidity and inhibit attachment at > 80% relative humidity; and that this attachment can occur even in the presence of high condensation nuclei concentrations.

DISCUSSION

The uptake of the airborne radioactivity by the blades of the fan could occur in two ways. First, since in the absence of the fan,

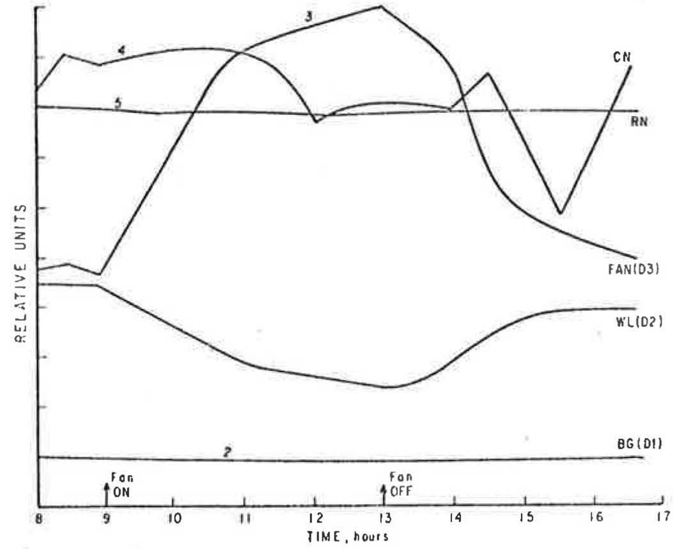

FIG. 4. Radon daughter concentration in the air (curve 1, full scale 10 WL/l), on the walls (curve 2, full scale 1000 counts/min), and on the fan (curve 3, full scale 3000 counts/30 min) in response to the switching on and off of the fan. Curve 4 (full scale $10^5/cm^3$), the condensation nuclei concentration, and curve 5 (full scale 2000 pCi/l.), the radon concentration, are practically independent of the fan being on or off. Humidity was <5%.

the daughter atoms seem to attach to the condensation nuclei present, raising the observed working level, the radioactivity could be transferred to the fan blades while attached to the particles. These particles are then collected onto the fan blades. Alternatively, the activity could remain unattached and move with the circulating air to the fan blades to which the active species becomes directly attached.

We suggest that the second mechanism provides the correct interpretation for these results. The RaA is most probably formed as a positive ion (Bi71). This ion can then be carried to the blades and becomes directly attached in preference to attaching itself to particles. The feasibility of this mechanism can be assessed by a theoretical calculation of deposition on a flat plate under conditions of turbulent flow. The deposition equation in a fully developing flat plate turbulent boundary layer is patterned after Fick's law but includes the effects of image charge forces (La78). In nondimensional units the equation is given as

$$(K/u^*)\left(1-\frac{y^+-S^+}{\delta^+-S^+}\right)=N_D\frac{\mathrm{d}C^+}{\mathrm{d}y^+}+N_{IM}\frac{C^+}{(y^+)^2}$$
$$=\text{deposition velocity} \tag{1}$$

where $K=J_0/C_\infty$; $y^+=yu^*/\nu$; $\delta^+=\delta u^*/\nu$; $S^+=Su^*/\nu$; $C^+=C/C_\infty$; $N_D=D/\nu+\epsilon/\nu$; $N_{IM}=[(\bar{\rho}_p{}^2a^5u^*)/54\mu\epsilon_0\nu^2]$ $(q/m)^2=(u^*q^2/96\pi^2\mu\epsilon_0\nu^2)$; J_0 = constant flux to surface; C_∞ = free stream concentration; $u^*=(C_f/2)^{1/2}U_\infty$ = friction velocity; C_f = flat-plate skin friction coefficient; ν = kinematic viscosity; y = coördinate normal to surface; δ = turbulent boundary layer thickness = $5.5\,(\nu x/U_\infty)^{1/2}$; x = coordinate parallel to and in the direction of the free stream velocity; U_∞ = free stream velocity; S = particle stopping distance = $2(0.99)\bar{\rho}_pa^2u^*/9\mu$; D = Brownian diffusivity; ϵ = turbulent eddy diffusivity; $\bar{\rho}_p$ = particle density; a = particle radius; ϵ_0 = permittivity of free space; μ = dynamic viscosity; q/m = charge-to-mass ratio of particle; and C = particle concentration. In order to solve this equation, it is necessary to know the functional relationships for $\epsilon(y^+)$. Lin *et al.* (Li53) suggest that there are three different boundary layers within the turbulent boundary layer: a laminar sublayer nearest to the plate surface, where $S^+\le y^+<5$; a middle region bounded by $5\le y^+<30$; and an outer region for which $30\le y^+\le\delta^+$. For the layer closest to the planar surface, $S^+\le y^+\le 5$, the relationship for ϵ is given by

$$\epsilon(y^+)=\nu(y^+/14.5)^3. \tag{2}$$

Substituting into equation (1) and rearranging yields

$$\frac{\mathrm{d}C^+}{\mathrm{d}y}=\left(\frac{K}{u^*}\right)\frac{1}{D/\nu+(y^+/14.5)^3}\left(1-\frac{y^+-S^+}{\delta^+-S^+}\right)$$
$$-\frac{N_{IM}}{D/\nu+(y^+/14.5)^3}\frac{C^+}{(y^+)^2}. \tag{3}$$

The diffusivity for the middle region bounded by $5\le y^+<30$ is

$$\epsilon(y^+)=\nu(y^+/5-0.959). \tag{4}$$

Substituting this expression into equation (1) would yield an expression similar to that given in equation (3) with the second term in the denominator altered for each of the two terms. Finally for the outer region of the boundary layer the diffusivity becomes

$$\epsilon(y^+)=\nu(0.098\delta^+-0.08y^+) \tag{5}$$

and again a modified version of equation (3) is obtained. In using these equations it is necessary to insure that there is a matching of boundary conditions at the interfaces between adjoining regions. A computer program has been written to solve the three deposition equations with the boundary conditions that

$$C^+=0 \quad \text{at} \quad y^+=S^+$$
$$C^+=1 \quad \text{at} \quad y^+=\delta^+$$
$$C^+\ \text{inner}=C^+\ \text{middle} \quad \text{at} \quad y^+=5$$
$$C^+\ \text{middle}=C^+\ \text{outer} \quad \text{at} \quad y^+=30. \tag{6}$$

The equations can be solved for various values of N_{IM}. Calculations indicate that the results are very weak functions of δ^+. These results can be applied to this experiment by considering the mass balance in the radon chamber.

$$Q_c(C_{in} - C_{out}) = J_0 A\alpha \tag{7}$$

where Q_c = flow rate through the chamber; C_{in}, C_{out} = concentrations for radon daughters in the inlet and the outlet, respectively; J_0 = deposition flux; A = active area of the fan = $(Q_f/Q_c)\cdot A_f$; A_f = actual surface area of the fan; Q_f = flow rate of air through the fan; and α = sticking coefficient. Then

$$\frac{C_{in} - C_{out}}{C_{in}} = \frac{J_0}{Q_c C_{in}} A\alpha \tag{8}$$

$$= \frac{(K)}{u^*}\frac{u^* A\alpha}{Q_c}. \tag{9}$$

Using our computer code and values for the parameters as follows: $u^* = 1.17 \times 10^2$ cm/sec; $Q_c = 1.21 \times 10^3$ cm^3/sec; $A = 1800$ cm^2; and D = diffusion coefficient of $RaA = 0.085$ cm^2/sec. K/u^* is calculated to be 0.041. Therefore,

$$\frac{(C_{in} - C_{out})}{C_{in}} = 7.1\alpha. \tag{10}$$

The actual reduction in working level is found to be 41%:

$$\frac{(C_{in} - C_{out})}{C_{in}} = 0.41. \tag{11}$$

For agreement between equations (10) and (11), the sticking coefficient, α, would have to be 0.058. This value is not an unreasonable estimate for *RaA* since it may exist as a molecular ion. As such, it would have a strong attraction for any surfaces with which it comes in contact. The deposition which has been described by these results could also be applied to the turbulent boundary layer associated with the wall instead of with the fan blades. However, the friction velocity is very much lower and there could not be the same high level of deposition as is associated with the fan blades.

Considering the analogous deposition of charged aerosol with its greatly increased mass and an anticipated much lower sticking coefficient, such a deposition is not likely. These particles are small enough ($<0.5\mu$) that impaction is not an important deposition mechanism. However, to insure that particles were not the mechanism of transport, the second time that the experiment was conducted, a search was made for deposited particles. Pieces of aluminum foil was cleaned and placed on the front and back of the fan blades. A piece was also attached to the wall of the chamber, and a fourth foil was kept as a blank to determine if particles were left after cleaning or deposited during storage and shipping. Each of these foils was then examined by scanning electron microscopy. The foils were coated with a thin conducting coat of gold so that good particle images could be obtained. The foils were then positioned in the scanning electron microscope (SEM) at a nominal magnification of 10^4. Examination was made of the field of view at regular intervals over each foil with 17–22 locations observed. For each location the number of particles was counted. The results of these analyses are given in Table 1. Considering that the number density of condensation nuclei was of the order of 10^5/cm^3, there is insufficient particle deposition onto the fan blades to account for the large drop in observed working level. In addition, since there are approximately as many *RaA* atoms as particles per unit volume, in order to have a 41% decrease in activity, there would have to be a discernible decrease in condensation nuclei. Thus, deposition of activity that is particle-attached can be eliminated from further consideration.

The mechanism proposed also provides an explanation of the observed effect of humidity on the fan deposition. At high humidity (>80%) the measured working level appears independent of the presence of the fan, indicating an inhibition of the uptake of daughter activity by the fan blades. The presence of the water molecules could be acting to

Table 1. Results of the microscopic examination for particle deposition

Foil	No. of Locations Observed	Total Area Viewed (μ^2)	No. of Particles	Particles / Unit Area
Front of fan blade	22	1251	72	$5.8 \times 10^6/cm^2$
Back of fan blade	17	907	18	$2.0 \times 10^6/cm^2$
Wall of Chamber	18	1023	13	$1.3 \times 10^6/cm^2$
Blank	22	1256	52	$4.1 \times 10^6/cm^2$

modify the nature of the *RaA* species and, thereby, substantially lower the sticking coefficient and also slow down the diffusion process. The water molecules could act as a scavenger for electrons ejected by the decaying radon atom from the path of ionization left by the emitted alpha particle. The negative water ion could then neutralize the positive *RaA* atom or ion molecule. It has been found by Wrenn (Wr77) that the presence of high humidity will partially neutralize *RaA* and prevent its deposition on the alpha-sensitive region of the New York University continuous radon monitoring instrument. The neutralization of the unattached activity will remove the effect of image charge in the diffusion through the boundary layer and will lower the sticking coefficient. Thus, the loss of the image charge force coupled with the reduction in sticking coefficient may be responsible for the observation of no appreciable deposition on the fan blades under high humidity conditions.

CONCLUSIONS

The experiments have shown that effects observed by Wrenn *et al.* (Wr69) and Shreve and Cleveland (Sh72) can be duplicated in a controlled laboratory environment and are then available for systematic study. The initial experiments have shown that radon daughter activity tends to plateout on the wall of the experimental chamber in the absence of condensation neclei and will remain airborne with high concentrations of small particle aerosol present. It has been shown that the high turbulence around a moving fan blade, coupled with the image force and high sticking coefficient, can cause substantial deposition of daughter activity onto the blades under conditions that permit the active species to retain a charge. The daughter transport to the blades in the chamber can be considered to consist of bulk transport to the fan blade area by the motion of the air caused by the fan and proximity transport of the activity through the boundary layer aided by image charge forces. Similar results regarding the role of image forces were found by Anderson *et al.* (An68). High humidity (>80% relative humidity) has been found to inhibit the deposition on the fan blade, presumably by providing a mechanism for neutralization of the charge on the active ion and thus lowering its sticking coefficient. Further studies are in progress to determine the detailed dependence of the rates of attachment and plateout of radon daughters on particle charge, composition and size, and number density as well as physical parameters such as humidity, temperature and flow conditions.

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