

# **Final Report**

July 2002 –2004

## **Experimental Investigation of Radio-Turbulence Induced Diffusion**

**AWARD # DE-FG07-02ID14340**

**6 September 2002 – 31 December 2004**

**By**

**Dr. Shoaib Usman (6 September 2003 – 31 August  
2004)**

**Dr. Henry Spitz (1 September 2004 – 31 December  
2004)**

**Nuclear and Radiological Engineering Program  
Department of Mechanical, Industrial and Nuclear  
Engineering  
University of Cincinnati  
598 Rhodes Hall  
Cincinnati, Ohio 45221-0072**

**1 July 2005**

# Experimental Investigation of Radio-Turbulence Induced Diffusion

## Final Report

## INDEX

Abstract	3
Introduction	4
Research Accomplishments	6
1 <sup>st</sup> Year	7
2 <sup>nd</sup> Year	11
Results	19
Conclusions & Future Research	22
References	25
Publications & Presentations	27

## Abstract

The outcome of this research project suggests that the transport of radon in water is significantly greater than that predicted solely by molecular diffusion. The original study was related to the long term storage of  $^{226}\text{Ra}$ -bearing sand at the DOE Fernald site and determining whether a barrier of water covering the sand would be effective in reducing the emanation of  $^{222}\text{Rn}$  from the sand. Initial observations before this study found the transport of radon in water to be greater than that predicted solely by molecular diffusion.

Fick's law on diffusion was used to model the transport of radon in water including the impact associated with radioactive decay. Initial measurements suggested that the deposition of energy in water associated with the radioactive decay process influences diffusion and enhances transport of radon. A multi-region, one-dimensional, steady-state transport model was used to analyze the movement of radon through a sequential column of air, water and air. An effective diffusion coefficient was determined by varying the thickness of the water column and measuring the time for transport of  $^{222}\text{Rn}$  through of the water barrier. A one-region, one-dimensional transient diffusion equation was developed to investigate the build up of radon at the end of the water column to the time when a steady-state, equilibrium condition was achieved. This build up with time is characteristic of the transport rate of radon in water and established the basis for estimating the effective diffusion coefficient for  $^{222}\text{Rn}$  in water. Several experiments were conducted using different types and physical arrangements of water barriers to examine how radon transport is influenced by the water barrier.

Results of our measurements confirm our theoretical analyses which suggest that convective forces other than pure molecular diffusion impact the transport of  $^{222}\text{Rn}$  through the water barrier. An effective diffusion coefficient is defined that includes effects of molecular diffusion and convection to describe the transport of radon in water. The effective diffusion coefficients measured in these experiments are  $6.8 \times 10^{-4} \pm 28\%$  and  $3.5 \times 10^{-4} \pm 34\% \text{ cm}^2 \text{ sec}^{-1}$  for the steady-state and transient diffusion conditions, respectively. Water barriers ranging in thickness from 30 – 50 cm reduce the amount of radon released from the radium-bearing source material by a factor of 0.3 – 0.1, respectively.

## Introduction

Diffusion coefficients,  $D_W$ , for radon are necessary to calculate particular values of the radon flux and the concentration profile. Values for the  $D_W$  largely depend on experimental measurements because no universal theory is available to perform accurate calculations, *a priori*. The radon diffusion coefficient in water cited by Adams [1964] is  $1.13 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$  at  $18^\circ\text{C}$ . This value was taken from Róna [1917] which was determined indirectly by applying the Einstein-Stoke equation. Lide [2003] quotes the  $D_W$  value given in Jähne et al. [1987], which also referenced the work by Róna [1917]. Figure 1 shows radon diffusion coefficients as a function of temperatures.

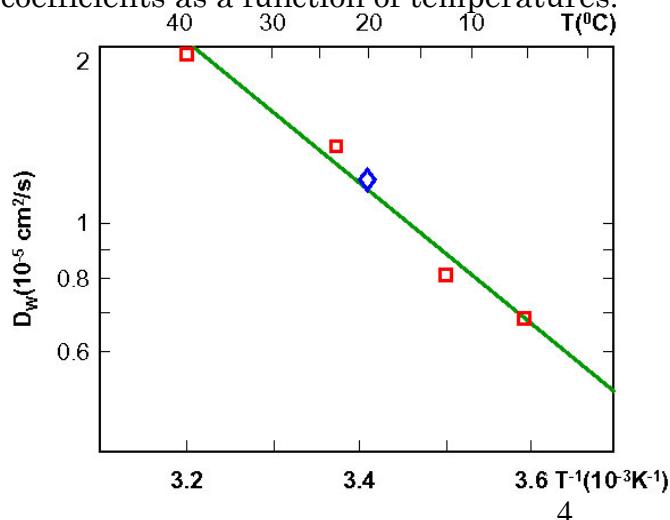


Figure 1: Diffusion coefficients of radon in water in  $\ln D$  against  $1/T$  representation. The diamond and square marks refer to data quoted in Róna (1917) and Hackbusch (1979), respectively, as cited by Jähne et al. (1987).

The methodology for determining  $D_w$  adopted in this research involved applying Fick's first law on diffusion to describe measurements of the concentration of  $^{222}\text{Rn}$  at the source and the exit (sink) end of a tubular water channel. This approach is similar to methods used to determine the diffusion coefficient of  $^{222}\text{Rn}$  in soil where the ratio of the  $^{222}\text{Rn}$  flux to the gradient concentration is a constant and consists of the  $^{222}\text{Rn}$  diffusion coefficient and soil properties, mainly the moisture content and porosity [Nielson, et al. 1982 ]. For soils, this constant is defined as the effective diffusion coefficient  $D_s$  and is on the order of  $10^{-2} \text{ cm}^2 \text{ s}^{-1}$  for low soil moisture and as low as  $2 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$  in a saturated soil.

Our research project was an outgrowth of a preliminary study to measure radon transport through a water barrier suggested as a means to reduce  $^{222}\text{Rn}$  emanation from two large concrete waste storage silos at the DOE Fernald site. These two silos formerly contained approximately  $6,126 \text{ m}^3$  of pitchblende ore residues that contain approximately 4,740 Ci of  $^{226}\text{Ra}$ , a significant source of  $^{222}\text{Rn}$ . Formerly, a fraction of the  $^{222}\text{Rn}$  would emanate from the silos and represented an exposure hazard for workers and the public.

The conventional mathematical model used to evaluate the transport of radon through a barrier of soil usually treats the problem as one dimensional steady-state molecular diffusion given by Fick's law. Cohen [1979] has evaluated methods for determining the diffusion coefficients by simple laboratory measurements. Silker [1981] adapted Cohen's method using a large scale radon attenuation test facility. Nielson et al. [1982] measured diffusion coefficients through the non-equilibrium or transient movement of radon through a sample material. Comparative measurements using air and soils showed good agreement with steady-state diffusion measurements.

## Research Accomplishments

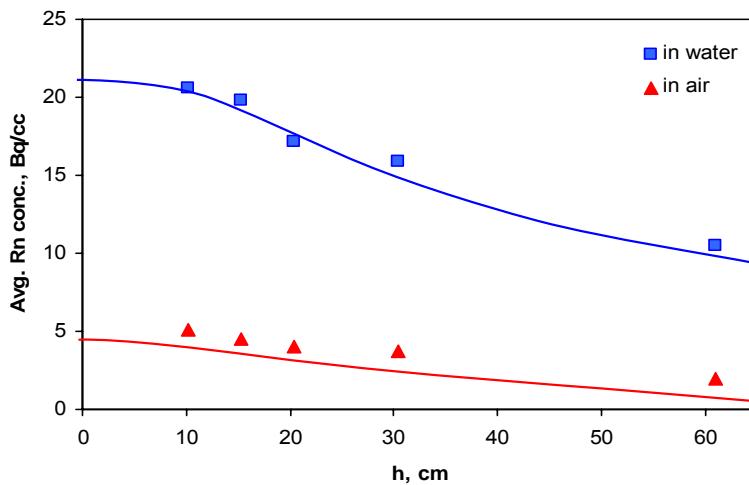
The main objective of the research was to describe the transport of radon in water, to predict what thickness of water barrier is required to obtain a certain level of attenuation of the radon flux, and to identify a source of convective energy that was observed to enhance transport beyond that predicted by molecular diffusion. A one-dimensional diffusion equation was first used to develop a description of the transport of radon in water. Measurements of the transport of  $^{222}\text{Rn}$  through still (i.e., turbulent-free) water barriers of several different lengths were performed to determine the effective  $^{222}\text{Rn}$  diffusion coefficient. The  $^{222}\text{Rn}$  that diffused through the tubular water barrier was derived from a constant activity source of  $^{226}\text{Ra}$ . The concentration of  $^{222}\text{Rn}$  at both ends of the tube was monitored for up to approximately 30 days. A one-dimensional diffusion equation with appropriate boundary conditions was adapted to describe  $^{222}\text{Rn}$  transport in still water for both steady-state and transient (i.e., time dependent) diffusion. One-dimensional, two-region (air and water) steady-state diffusion equations were also used to determine the  $^{222}\text{Rn}$  effective diffusion coefficient,  $D_W$ , in water. The transient diffusion equation solution provides a comparison for the steady state calculation and evaluates the diffusion process in greater detail.

Samples of the radium bearing waste stored at the Fernald Environmental Management Project (FEMP) were formerly evaluated at the University of Cincinnati for  $^{226}\text{Ra}$  content, radon emanation fraction, and the reduction in emanation created by different thicknesses of water covering the waste [Spitz 1999]. The  $^{226}\text{Ra}$  content of the samples of K-65 waste material evaluated at UC was  $(13.74 \pm 0.3)$  kBq g $^{-1}$ . Five vertical PVC columns having a diameter of 6 inches and increasing lengths were constructed. At the bottom of each column was approximately 40 g of the

K-65 radium-bearing waste material sealed between two fiberglass filters. The PVC columns were filled with water to cover the radium-bearing waste to a depth of 10, 15, 20, 30, and 61 cm, respectively. The air volume above each water barrier was fixed and equal to 18.1 L. The radon concentration in the water was sampled at different depths using a 1 mL gas-tight syringe and measured for radon content by mixing with 15 mL of liquid scintillator.

#### First Year: Effective Radon Diffusion Coefficient, $D_w$

The radon concentrations in air and water for different water thicknesses are shown in Figure 2.



**Fig 2: Avg. radon conc. in water and air at steady state, where  $h$  is the thickness of water column.**

As expected, the radon concentration in water varies with the total water volume. The combined effects of transport time and radioactive decay explain the change of radon in air above the water barrier with the barrier thickness.

Fick's law, a one-dimensional, steady-state effective diffusion equation was used to model the transport of radon in the water. An effective diffusion coefficient for radon in water,  $D_w$ , was adopted that includes all external forces that may impact transport. These external

forces cause convection resulting in an effective diffusion coefficient  $D_W$ , which is larger than the pure molecular diffusion coefficient for radon. The value of  $D_W$  is specific for these experimental circumstances,  $J$  is the flux of radon passing  $x$  and  $C$  is the free radon concentration at  $x$  and  $\lambda$  is the radon decay rate.

$$\frac{d^2C}{dx^2} = \frac{\lambda}{D_W} C$$

The general solution to this diffusion equation is  $C(x) = P e^{kx} + Q e^{-kx}$

where,  $k = \sqrt{\frac{D_W}{\lambda}}$ ,  $P$  and  $Q$  are constants that are determined by the boundary conditions of the system, i.e.,

$$C(0) = C_0 \text{ and } J(h) = 0 \text{ or } \left. \frac{dC}{dx} \right|_{x=h} = 0 .$$

Applying these boundary conditions, the solution for radon concentration is

$$C(x) = C_0 \frac{e^{2kh} + e^{2kx}}{1 + e^{2kh}} e^{-kx} .$$

The concentration of radon at the water surface is  $C_h = C_0 \operatorname{Sech}(h \sqrt{\frac{\lambda}{D_W}})$ .

Newton's method was used to obtain  $D_W$  for measured values of  $C_h$  at  $h$ .

The expression for  $D_W$  was also determined from the average  $^{222}\text{Rn}$  concentration measured in the air above each water column thickness at equilibrium or  $C_h(\text{air})$ . The value of  $C_h$  was determined using Henry's Law,

$$C_h = \frac{C_h(\text{air})}{H}$$

where  $H$  is Henry's law constant<sup>1</sup> for Rn gas in water. The value of  $H$  can be expressed as the ratio of Rn gas concentrations at the air-water interface. At steady-state, the activity concentration (Bq/cc) for radon in air and water was also used to determine  $H$ .

A two-region diffusion equation was developed using the conditions shown in Figure 3:

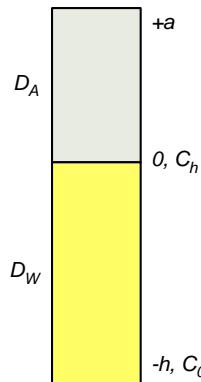


Figure 3: Two-region, steady state diffusion model.

The 'initial' radon concentration,  $C_0$ , is transported through the water thickness  $h$  with effective radon diffusion coefficient  $D_W$ . The release of radon into the air column through height  $a$  is also treated as a diffusion process with diffusion coefficient  $D_A$ . The radon concentration at the interface is  $C_h$ , measured directly above the water surface.

The solution to the diffusion differential equations for the water at the interface ( $x = 0$ ) is

---

<sup>1</sup> Henry's law states the concentration of a gas in a liquid ( $C$ ) is directly proportional to the partial pressure of the gas above the surface of the liquid ( $P_g$ ), or  $C = k_H P_g$ , where  $k_H$  is Henry's Law constant. In dimensionless unit the constant can be expressed as  $H = C_g/C$ , where  $C_g$  is the gas concentration above the liquid.

$$C_h = \frac{C_0 k_A}{k_A \cosh(k_w h) + H k_w \sinh(k_w h) \operatorname{Tanh}(k_A a)}$$

where

$$k_w = \sqrt{\frac{\lambda}{D_w}} \text{ and } k_A = \sqrt{\frac{\lambda}{D_A}}.$$

All the diffusion coefficients calculated using the one-dimensional steady-state diffusion equation were two orders of magnitude greater than the reference value, i.e.  $1.13 \times 10^{-5} \text{ cm}^2 \text{ sec}^{-1}$ , which suggests the presence of convective forces in addition to diffusion. In this approximation, all mixing caused by the sampling process is averaged out as a single steady convection to fulfill to the concept of an effective rate of movement of radon.

The predicted and experimental results were equivocal, although it is recognized that external convective forces arising from the method of sampling caused radon transport to be greater than that produced solely by molecular diffusion. The value for  $D_w$  obtained using the Henry's law constant is almost the same as the value obtained directly using the one-region approach and validates the model based upon the Henry's law constant. This constant is also required in the two-region steady-state diffusion approach.

The  $^{222}\text{Rn}$  partial pressures above the water were also relatively small. So it is relevant to compare the Henry's law constant obtained in this experiment (viz., 0.23) to that of a reference at standard conditions ( $20^\circ\text{C}$  and 1 atm.) which is 0.277 [Error! Reference source not found.]. The average ambient temperature during the whole experiments was relatively constant ( $20 \pm 3$ ) $^\circ\text{C}$ .

## Second Year: Steady-State and Transient Diffusion

### Steady State Diffusion

The difference in  $D_w$ 's obtained using the one- to two-region model is significant for large air to water column ratios. Using the value of  $D_w$  from the two-region model as a reference value, a  $D_w$  correction factor

$$\Delta D_w \equiv \frac{|D_w - D_w^1|}{D_w}$$

was calculated, where  $D_w^1$  is the diffusion coefficient calculated using one-region model.

Henry's law breaks down when partial pressures for  $^{222}\text{Rn}$  exceed about 500 kPa and/or when the dissolved concentration exceeds 1% (Poling et al. 2001). However, radon partial pressures and fractions in the water and in the headspace encountered in this project were very small which justifies use of a constant value of  $D_w$  in the diffusion differential equation.

Three water channels having a circular cross section and increasing lengths were designed and fabricated. This design was selected because it was easy to fabricate and minimized any abrupt wall effects. A constant source of radon is attached at one end of the channel. Measurements of radon at the source and terminal ends of the channel are performed as the means of quantifying transport. The radon source is connected to one end (source) of the tube. Radon laden air is measured on the source and sink ends of the tube. Transient diffusion of radon occurs at the onset when the

source is first connected to the water. The radon concentration increases with time in the water and at the sink end of the water column. Once radon reaches its maximum concentration at the sink, steady-state diffusion will continue and the rate of diffusion into the sink air space is equal to the rate of radioactive decay.

Three geometrical arrangements for the cylindrical diffusion channels were investigated.

- a) vertical diffusion channel with the radon source located above the water column,
- b) horizontal diffusion channel with the radon source place at an end, and
- c) a U-tube arrangement.

The transient and steady-state transport of radon in water were evaluated using measurements of  $^{222}\text{Rn}$  in air and water. At the end of the experiment, when radon in air achieved a steady-state concentration, samples of air were extracted from the space above the water surface and water was removed using a 1 ml gas-tight syringe at the air/water boundary. Alpha activity in each sample was counted in the liquid scintillation counter.

A three-region, one-dimensional, steady-state diffusion equation was used to model the transport of radon in water at steady-state for the physical conditions presented in the channel experiments. This is a more generic setup which can also be implemented to the two-region conditions. The steady-state differential equations for radon diffusion in water and air at both ends of the water channel, respectively, are

$$D_A \frac{d^2 C_1}{dx^2} - \lambda C_1 = 0$$

$$D_w \frac{d^2 C_w}{dx^2} - \lambda C_w = 0$$

$$D_A \frac{d^2 C_2}{dx^2} - \lambda C_2 = 0$$

where  $C_1$ ,  $C_w$  and  $C_2$  refer to radon concentration at location  $x$  in the air at the source side, in the water and in the air at the sink side with the diffusion coefficient  $D_w$  (water) and  $D_A$  (air); and  $\lambda$  is radon decay constant.

The solution to the three-region steady state diffusion equations for the water at the interface ( $x = 0$ ) is

$$C_h = \frac{C_0 k_A}{k_A \cosh(k_w h) + H k_w \sinh(k_w h) \operatorname{Tanh}(k_A a)}$$

where  $k_w = \sqrt{\frac{\lambda}{D_w}}$  and  $k_A = \sqrt{\frac{\lambda}{D_A}}$ .

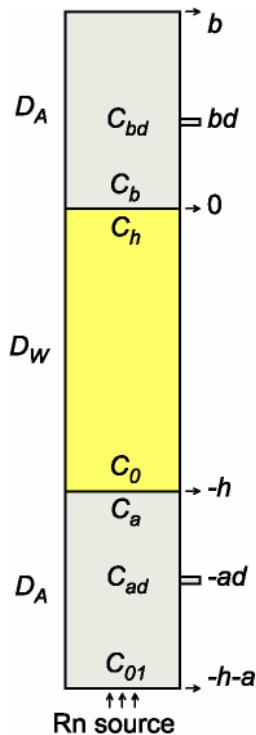


Figure 4: Three-region steady-state diffusion model.

If the known parameters are  $C_b$  and  $C_a$  instead of  $C_h$  and  $C_o$ , the expression becomes

$$C_b = \frac{C_a H^2 k_A}{k_A \text{Cosh}(k_W h) + H k_W \text{Sinh}(k_W h) \text{Tanh}(k_A b)}$$

Using the air concentration measurements in the source and sink,  $C_{bd}$  and  $C_{ad}$ , respectively, at some distance from each interface, the solution to the three-region diffusion equation becomes

$$C_{bd} = 2 C_{ad} H^2 k_A k_W e^{k_W h} \text{Cosh}\{(b-bd)k_A\} / \\ [\{H k_A \text{Sinh}(k_A ad) - k_W \text{Cosh}(k_A ad)\} [H k_W \text{Sinh}(k_A b) - k_A \text{Cosh}(k_A b)\} + \\ \{e^{2k_W h} [k_W \text{Cosh}(k_A d) + H k_A \text{Sinh}(k_A ad)] [k_A \text{Cosh}(k_A b) + H k_W \text{Sinh}(k_A b)]\}.$$

### Transient diffusion

Since radon transport in water is small compared to transport in soil, the method developed by Nielson et. al [1982] was adopted for this project. The one-region, one-dimensional, time-dependent radon diffusion equation is

$$D_W \frac{\partial^2 C}{\partial x^2} - \lambda_1 C = \frac{\partial C}{\partial t}.$$

where  $C$ : radon concentration in water at distance  $x$  and time  $t$  of the water column,

$D_W$ : radon diffusion coefficient in water,

$\lambda_1$ : radon decay constant.

Defining the origin as the source end of the diffusion column, the following boundary conditions were used:

$$C(x,0) = 0 \quad \text{and} \quad C(0,t) = C_0 \quad \frac{\partial C}{\partial x} \bigg|_{\substack{x=h \\ \text{all } t}} = 0$$

Taking the Laplace transform with respect to  $t$  and applying the preceding boundary conditions, the solution derived by Nielson [1982] is

$$C_h(t) = C_0 \sum_{n=1}^{\infty} \frac{(-1)^{n-1} (2n-1)}{\pi (2n-1)^2 / 4 + \lambda_1 h^2 / (\pi D_W)} \left( 1 - e^{-\lambda_1 t - (2n-1)^2 \frac{\pi^2 D}{4h^2} t} \right)$$

where  $C_h$ : radon concentration in water at the exit end of the water column in the sink side,

$C_0$ : radon concentration in water at the entrance of the water column in the source side,

$h$ : water column length,

$D_W$ : radon diffusion coefficient in water,

$\lambda_1$ : radon decay constant

This solution was the basis for interpreting the transient diffusion experimental data. Two modifications to the theory developed by Nielson were introduced. The first modification incorporates the Bateman equations to account for contributions from the radon decay products,  $^{218}\text{Po}$  and  $^{214}\text{Po}$  [0]. The second modification was to adopt a two-region transient diffusion solution. A third modification unique to this research involves incorporating the Henry's law constant to determine the concentration of  $^{222}\text{Rn}$  in water from the measured concentration of  $^{222}\text{Rn}$  in air at the sink end of the column. This modification was possible because  $C_0$  is a constant and only  $C_h$  at steady-state is required.

Figure 5 shows the theoretical relative radon concentration at the sink with time for increasing values of the diffusion coefficients in water,  $D_W$ . The value of  $D_W$  determines the rate of  $^{222}\text{Rn}$  transfer in the water and the resulting attenuation ( $C_h/C_0$ ) at steady state. Experimental results for  $C_h$  at various times will be compared to these theoretical curves.

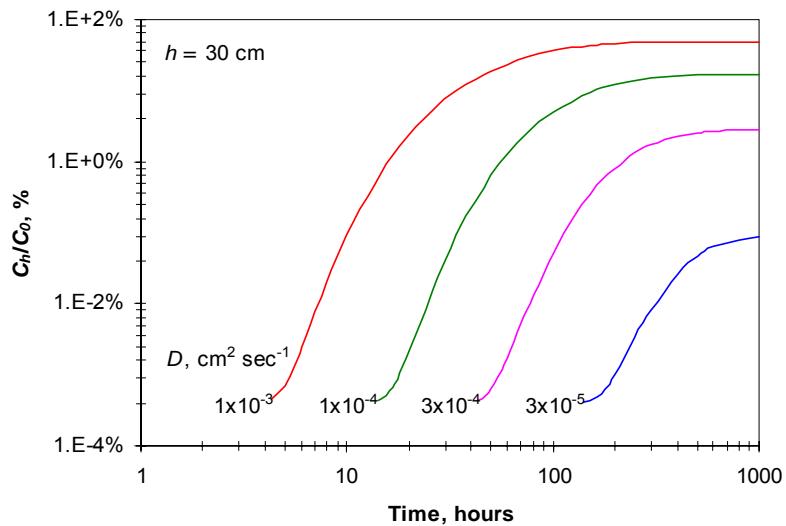


Figure 5: Predicted transient diffusion at the end of 30 cm water column for different diffusion coefficients

Since the two-region transient diffusion problem was very complex and difficult to solve, a correction factor was derived using the ratio of the radon concentration predicted by the steady-state two-region water and air problem ( $C_{2R}$ ) to that predicted by the steady-state one-region water problem ( $C_{1R}$ ). These two concentrations were derived previously. After simplification, the two-region correction factor becomes

$$\frac{C_{2R}}{C_{1R}} = \left[ 1 + \sqrt{\frac{D_A}{D_W}} \tanh(k_w h) \tanh(k_A a) \right]^{-1}.$$

This factor gives an exact correction for the final plateau region of the transient curves, and is multiplied by the source concentration.

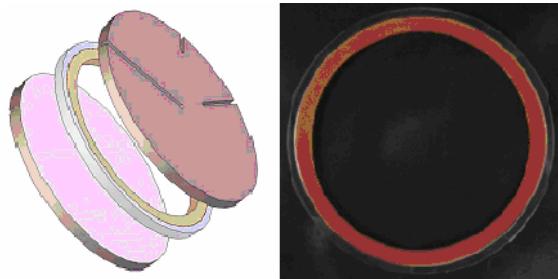
## Second Year: Bench-marking for Heat Transfer Experiment

Our study of the transport of  $^{222}\text{Rn}$  through water is similar to the standard transport of a scalar quantity through an incompressible fluid with special attention directed toward the effect of alpha radiation on the phenomenon of mass transfer. Due to similarities between mass and heat transfer we initiated a study to investigate the effect of radiation on heat transfer. Heat transfer enhancement due to radiation is to be investigated by using Raleigh-Bénard [RB] convection experiments and numerical simulations. RB convection experiments are standard means of investigating natural convection phenomenon. The simple experiment is made up of two parallel horizontal plates. Fluid is enclosed between the two plates. Bottom plate is heated and the on-set of convection is studied. For sufficiently large parallel plates, the transition to convective heat transfer mode is expressed as a critical value of Rayleigh number (Bejan 2004), which is defined as;

$$Ra_H = \frac{g \cdot \beta \cdot (T_h - T_c) \cdot H^3}{\alpha \cdot v}$$

where,  $g$  is acceleration due to gravity,  $\beta$  is the coefficient of volume expansion  $T_h$  and  $T_c$  are the hot and cold temperatures respectively.  $H$  is the height or distance between the two plates and  $\alpha$  is the thermal diffusivity, while  $v$  is the viscosity of the fluid. For a sufficiently large size plates, convective cells are observed immediately above  $Ra \geq 1708$ . Bénard cells experiments are very repeatable and are used to demonstrate transition of the mode of heat transfer under different conditions.

We have fabricated a prototype Benard cell and collected benchmark data. Polished copper was used to manufacture two circular plates of 9.22 cm in diameter. A circular geometry was adopted to eliminate any local convection currents. A layer of fluid was held between the plates with the help of a 4.5 mm pixy glass spacer and circular seal (Figure 6). Thickness of the spacer was calculated to ensure that for the test temperatures convection would be the mode of heat transfer.



**Figure 6: Raleigh-Bénard Apparatus (L), Seal & Spacer (R)**

Three 44000 thermistors were inserted in the groves on each plate to collect temperature data. In view of the slow response and high precision required for this application thermistors were found to be the best temperature sensors (Scarangella 2003i). Three temperature measurements were averaged to maintain best accuracy. Data were collected using National Instrument High-Precision Temperature and Voltage Logger data acquisition card. A simple temperature regulator circuit was designed to control the bottom plate temperature within a fraction of a degree Celsius.

The apparatus was mounted between two insulation ceramic layers to minimize heat loss from the system. The perimeter edge was insulated using polystyrene foam.

The Raleigh-Bénard convection apparatus was loaded with the test fluid and positioned horizontally. Figure 7 shows the final apparatus enclosed in a polystyrene insulator.



Figure 7: Test apparatus

Measurements using this test apparatus resulted in development of a useful analogy model for convective heat transfer (Usman et. al. 2005). Additional work will include alpha convection. These experiments will remove the possible interference between the phenomenon of scalar transport and the source of radiation which was the same for radon experiment.

## Results

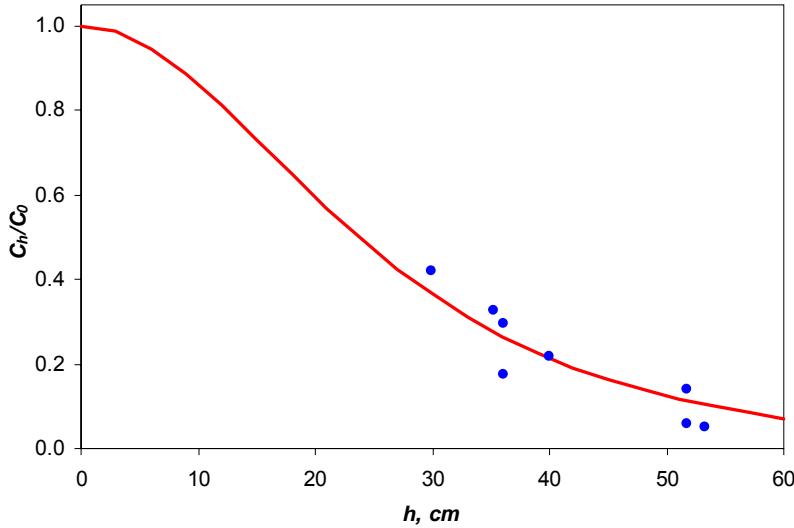
### Steady-State Diffusion

The results of determining  $D_W$  for the two- and one region models are listed on

$h$ , cm	$C_o$ , Bq/cc	$C_h/C_o$	$D_W$ , $\text{cm}^2 \text{ s}^{-1}$	
			Two Reg	One Reg
30	4	0.42	8.9E-04	8.2E-04
35	15	0.33	8.6E-04	8.2E-04
36	3	0.18	4.8E-04	4.7E-04
36	1	0.30	7.9E-04	7.7E-04
40	2	0.22	7.0E-04	7.0E-04
52	177	0.06	4.6E-04	4.6E-04
52	44	0.14	8.0E-04	7.9E-04
53	2	0.05	4.5E-04	4.4E-04
Averaged $D$ , $\text{cm}^2 \text{ s}^{-1}$			6.8E-04	6.6E-04
$1\sigma$ error			28%	26%

Table 1: Radon diffusion coefficients in water determined from steady-state measurements.

shows the attenuation curve of radon in water using the two-region model and an average value of  $D_W$  equal to  $6.8 \times 10^{-4} \pm 28\% \text{ cm}^2 \text{ s}^{-1}$ .



**Figure 6:**  
Observed and predicted attenuation of radon in water for different length of water column for  $D_W = 6.8 \times 10^{-4} \text{ cm}^2 \text{ sec}^{-1}$ .

Figure 6 shows a good agreement between  $^{222}\text{Rn}$  attenuation and the length of the water column with the model. Furthermore, there is no correlation between  $^{222}\text{Rn}$  concentration and  $D_W$ , which validates the assumption that  $D_W$  is constant for the whole experiment. Likewise, there is no correlation of  $D_W$  with tube diameter (16 - 25 mm) or tube geometry (U-tube and horizontal columns). However, there may be a correlation between  $D_W$  and the ratio of water tube diameter and length since no measurements were conducted with  $h < 30$  cm. The use of one-dimension in the diffusion method requires a small ratio to eliminate effects in the lateral diffusion. The results from one- and two-region models do not show significant differences because the length of the air columns,  $a$ , in the second region are relatively small, i.e. between 1 to 6 cm.

The value of  $D_W$  determined from these experiments is approximately 55 greater than the conventional value. Further research is being conducted to examine the microturbulence produced by  $\alpha$ -particles from

the decay of radon and its progeny that may affect the radon transport rate in the water.

### Transient Diffusion

Data from the transient diffusion experiments were fit to the predicted curve obtained by solving the transient diffusion equation. Table 2 compares values for  $D_w$  derived from the steady-state and transient  $^{222}\text{Rn}$  diffusion models. Again, the attenuation of the  $^{222}\text{Rn}$  concentration in the water column is adequately described using the two-region model with the transient diffusion equation and an average value for  $D_w$ . However, the value for  $D_w$  predicted using the steady-state diffusion model is always greater by than that predicted using the transient diffusion model. This difference is likely related to systematic experimental error and identifying the time at which a steady-state condition is established.

$h$ , cm	$C_0$ , Bq/cc	$C_h/C_0$	$D_w$ , $\text{cm}^2 \text{ sec}^{-1}$	
			Steady-state	Transient
19	7.4	0.32	2.95E-04	1.90E-04
30	4.2	0.42	8.94E-04	4.94E-04
35	15.2	0.33	8.56E-04	4.69E-04
36	1.1	0.30	7.93E-04	3.47E-04
52	177	0.06	4.61E-04	3.66E-04
53	2.3	0.05	4.47E-04	2.44E-04
Averaged $D_w$ , $\text{cm}^2 \text{ s}^{-1}$			6.24E-04	3.52E-04
$1\sigma$ error			41%	34%

Table 2: Comparison of  $D_w$  determined from steady-state and transient of  $^{222}\text{Rn}$  concentrations.

The curve describing transient diffusion of  $^{222}\text{Rn}$  in water will eventually reach a steady-state condition where the  $^{222}\text{Rn}$  concentration is constant with time. The time necessary to achieve a steady-state

condition,  $t_{SS}$ , depends on the water column length,  $h$ , and the diffusion coefficient,  $D_w$ . At very large  $t_{SS}$ , the slope of the curve is very small, which makes it difficult to uniquely identify the time when a steady state condition is achieved. The 90%  $t_{SS}$  value is easier to determine and was calculated with  $t = \infty$  to obtain the  $^{222}\text{Rn}$  concentration at steady state,  $C_{SS}$ . The 90% concentration,  $C_{90}$  is equal to 0.9  $C_{SS}$ . (Figure 7) The Newton method was used to determine the time to achieve  $C_{90}$ , i.e. 90%  $t_{SS}$ .

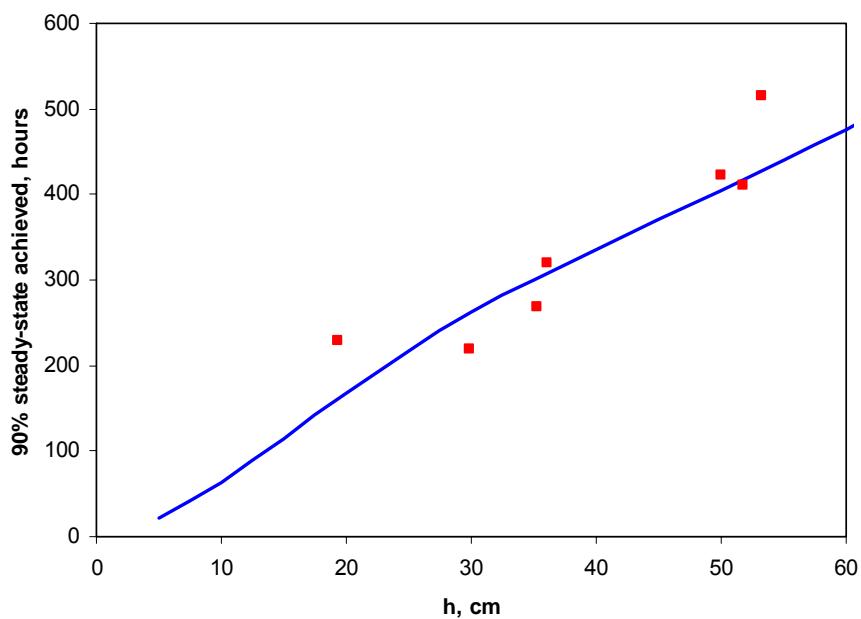


Figure 7: 90% steady-state time against water length of the  $D_w$  measured and the transient

### Conclusions & Future Research

Experiments have demonstrated that the water columns are effective in reducing the  $^{222}\text{Rn}$  concentration in the sink air and that Henry's law could be used to determine the radon concentration at the end of the water column from measurements of  $^{222}\text{Rn}$  in sink air. Measurements of  $^{222}\text{Rn}$  in water columns of increasing length are consistent with predictions using the one-dimensional steady-state diffusion model from which the effective diffusion coefficient for  $^{222}\text{Rn}$  in water can be determined. The effective

diffusion coefficient reflects the transport of  $^{222}\text{Rn}$  in water by molecular diffusion and convective processes. The strong correlation between the predicted and measured concentration validates the model.

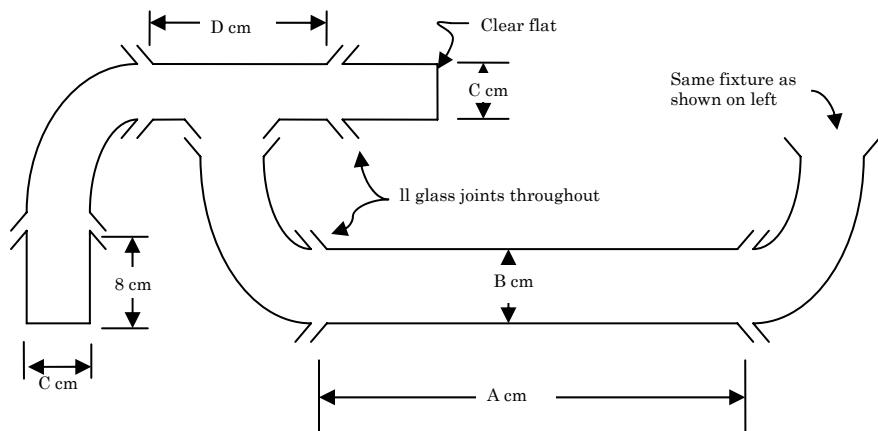
The diffusion coefficients,  $D_W$ , obtained using Henry's law to determine the concentration of  $^{222}\text{Rn}$  in water is approximately equal to the  $D_W$  obtained by direct measurement of  $^{222}\text{Rn}$  at both ends of the water column. Henry's law has been implemented in diffusion model described in this thesis. The Henry's law constant for radon determined by direct measurement in this research is  $0.21 \pm 14\%$  and is supported by the other experimental results.

The diffusion coefficient for  $^{222}\text{Rn}$  in water determined using the steady-state diffusion model was equal to  $6.8 \times 10^{-4} \pm 28\% \text{ cm}^{-2} \text{ sec}^{-1}$ . The transient diffusion model leads to a diffusion coefficient equal to  $3.5 \times 10^{-4} \pm 34\% \text{ cm}^{-2} \text{ sec}^{-1}$ . These values are independent of the activity of the source of  $^{222}\text{Rn}$  in the range of 1 - 176 kBq/cc and the length of the water channel in the range 30 – 53 cm. Radon-222 emanation is reduced from 10% up to 36% for water channel lengths ranging from 30 cm to 53 cm.

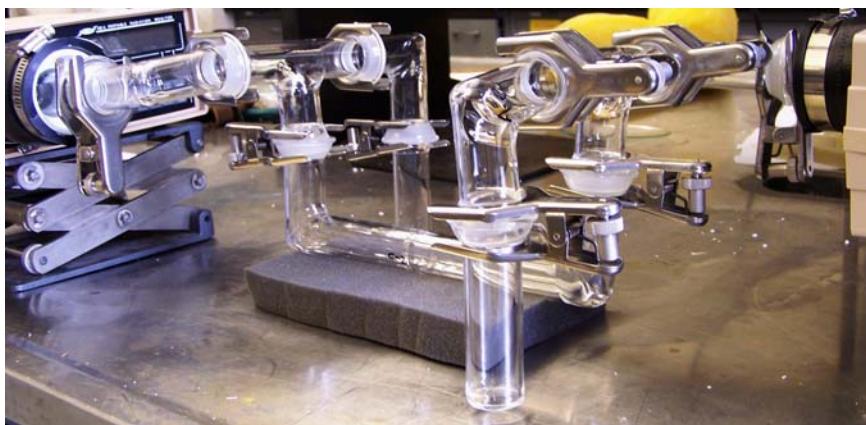
The effective diffusion coefficient determined in this research is a result of pure molecular diffusion plus other sources of convective energy. The pure molecular diffusion coefficient for  $^{222}\text{Rn}$  cited in the literature ranges from  $1.14 \times 10^{-5}$  to  $1.56 \times 10^{-5} \text{ cm}^{-2} \text{ sec}^{-1}$ . The effective diffusion coefficients determined in this research reflects the average rate of the total transport mechanisms of radon in the water.

Follow-up research is being conducted to measure radon transport in a fixed, isolated system that eliminates all external forces that could introduce turbulence and influence transport. The system involves a tubular horizontal glass diffusion column. (Figures 8 & 9) Continuous measurements of  $^{222}\text{Rn}$  at the source and sink ends of the column are

performed using ZnS(Ag) coated glass tubes in contact with photomultiplier tubes. Analysis of measurements will be conducted using the transient diffusion model.



**Figure 8:**  
Schematic drawing of rigid glass diffusion apparatus for measuring radon transport in water



**Figure 9:**  
Experimental arrangement of the rigid glass diffusion apparatus connected to

## References

Adams, J.A.S., Lowder, W.M., "The Natural Radiation Environment, University of Chicago Press, 1964.

Bejan, A. *Convective Heat Transfer*, John Wiley & Sons, New York, 2004.

Cohen, B.L., "Methods for Predicting the Effectiveness of Uranium Mill Tailing Covers", Nucl. Inst. and Meth. 164 (1979), p. 595-599.

Hawwari, M. "Investigation and Simulation of Raleigh-Bénard Convection in a cylindrical Geometry", M.S. Project, College of Engineering, University of Cincinnati, 2004.

Jähne, B., Heinz, G., Dietrich W., "Measurements of the Diffusion Coefficients of Sparingly Soluble Gases in Water", J. Geophys. Res., 92, 10767, 1987.

Lide, D.R., Ed-in-Chief, "CRC Handbook of Chemistry and Physics, 84th ed., 2003-2004.

Nielson, K.K., Rich, P.C., Rogers, V.C., "Comparison of Radon Diffusion Coefficient Measured by Transient-Diffusion and Steady-State Laboratory Methods", NUREG/CR 2875, U.S. Nuclear Regulatory Commission, 1982.

Poling, B.E., Prausnitz, J.M., O'Connell, J.P., "The Properties of Gases and Liquids", 5th Edition, 2001.

Róna, E., "Diffusiongrösse und Atomdurchmesser der Radium-emission", ("Magnitude of diffusion and atomic dimensions of radium emanation"), Ztschr. phys. Chemie., 92: 213-18. (1917).

Scarangella, M. "Benchmarking of Bénard Cell Convection Apparatus", M.S. Project, College of Engineering, University of Cincinnati, 2003.

Silker, W.B., A Radon Attenuation Test Facility", NUREG/CR 2243, U.S. Nuclear Regulatory Commission, 1981.

## Publications and Presentations

H. Spitz, S. Usman, S. Syahrir. The effect of radio-turbulence on the transport of radon in water. Manuscript in Progress.

S. Syahrir, S. Usman, H. Spitz, J. Weisman. Transport of radon in still water under steady state and transient conditions. 11<sup>th</sup> International Congress of the International Radiation Protection Association. Madrid, Spain. May, 2004.

S. Usman, S. Syahrir, H. Spitz, J. Weisman. Transport of radon in still water. Council on Ionizing Radiation and Measurements. National Institute for Standards and Technology. Bethesda, Maryland. October 2004.

H. Spitz, S. Syahrir, S. Usman, J. Weisman. Transport of radon in water under steady-state and transient conditions. 49<sup>th</sup> Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry. Jackson Hole, Wyoming. 2003.

---