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RADON PROGENY CONTROL IN BUILDINGS

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FOREWORD

This report was prepared by Michel V. J. Culot as a dissertation in partial fulfillment of the requirements for the degree of Doctor of Philosophy. Mr. Culot has been involved with this research project since its inception in 1970, and has been responsible for much of the progress made to date.

Many questions related to the evaluation and reduction of radiation exposures in structures where uranium mill tailings were used in construction remain unanswered. Although this dissertation contains information that can be extremely helpful in the Grand Junction remedial action program, it should be considered merely as a progress report in relationship to the total needs of the program. However, to the agencies that supported only these initial portions of the research, this dissertation is presented as a final report.

K. J. Schiager

ABSTRACT

RADON PROGENY CONTROL IN BUILDINGS

The use of Uranium mill tailings as a construction material around habitable structures has resulted in augmented whole body gamma exposures and in increased Radon progeny lung exposures to the occupants. The use of sealants designed to prevent Radon diffusion into the structures is investigated for application in the most frequently encountered case, i.e. where the gamma exposure levels do not warrant the costly removal of the tailings but where the Radon progeny concentration exceeds the standards set for the general public.

The fractional increase in whole body gamma exposure resulting from the application of Radon barriers on interior surfaces of a structure is analyzed. A review of the parameters affecting the diffusion of Radon in concrete, and a method to evaluate the effective diffusion coefficient of a concrete slab, are presented. The modifications introduced in the Radon concentration profiles by a Radon barrier are evaluated by application of a linear diffusion model to a multilayered system of porous media. The prediction of the resulting increase in whole body gamma exposures is obtained for various practical cases by the introduction of the original and modified concentration profiles in shielding calculations involving infinite slabs with variable radiation volume source strengths.

The results of a testing program which led to the identification of an effective Radon sealant are presented with the experimental evidence demonstrating the effectiveness of the sealant by the successful

reduction of the average Radon concentration following its application
in an experimental building.

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We wish to acknowledge the help of the numerous persons which, in one way or another, have contributed to the success of this interdisciplinary study. The advice of our Professor, Dr. H.G. Olson (Mechanical Engineering Department) in directing our work and his constant availability in times of discouragement were of irreplaceable value. We wish to thank especially Dr. K.J. Schiager (Department of Radiology and Radiation Biology) for his numerous teachings in the art of radiation measurements and in the field of radiation health physics; the present work would have not reached completion without his support. The interest showed in our work by Dr. R.D. Haberstroh (Mechanical Engineering Department) stimulated the deepening of thoughts which was helpful in many aspects of the study. The mathematical expertise of Dr. H.E. Wilhelm (Physics Department), R.B. Kelman (Department of Mathematics and Computer Science), F.W. Smith (Mechanical Engineering Department) was invaluable in resolving preliminary problems not reported in this work but which allowed a better understanding of the difficulties involved.

The help and information sharing of Dr. Schiager's doctoral student, D.R. Alexander facilitated enormously the difficult problem of instrumentation and the skills of K. Wilson in the fabrication of Radon Scintillation chambers contributed a great deal to the success of the measurements. The information provided by Drs. J.R. Goodman and M.D. Vanderbilt (Civil Engineering Department) on concretes and Dr. D.K. Sumada (Civil Engineering Department) on the diffusion processes in porous media, was of importance in starting our research effort. We thank Dr. Brown (Ideal Cement Co., Fort Collins, Colorado) for his

interest and his permission to use the cement plant's specialized library on concretes. The assistance of our fellow graduate student, J.E. Koske, in recording data and performing the heavy manual work which was necessary in certain aspects of the study was deeply appreciated.

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LIST OF SYMBOLS AND ABBREVIATIONS

Symbol

A_{ij}, A_{ij}^*	j^{th} coefficient in expression of i^{th} population in a radioactive chain (atoms)
A_{1i}, A_{2i}	Weighing factors for experimental fit of buildup
A	Surface area covered by sampling container (dm^2)
a	Rate of production of Radon within the soil ($\text{at}/\text{cm}^3\text{-s}$)
$\text{\AA}$	Angstrom (10^{-8}cm)
A_o	Activity of Radon sample in Radon scintillation chamber at the time of sampling (c/time)
$A(t)$	Activity of Radon sample in Radon scintillation chamber at the time of counting
AEC	Atomic Energy Commission
AM-9	Chemical grout
AP	Amonium persulfate
at	Atom
B	Buildup factor
B_{ij}^*	j^{th} coefficient in expression of i^{th} population in a radioactive chain (atoms)
BaCl_2	Barium Chloride
Bi-214	Isotope 214 of element Bismuth
BNL	Brookhaven National Laboratory
c	Count
$[C]_o$	Initial CCl_4 concentration (ppm)
$[C]_t$	CCl_4 concentration at time t
C_{14}	Radon concentration in the gas phase of the tailings before the application of a Radon barrier (pCi/cm^3)
C_{24}	Radon concentration in the gas phase of concrete before the application of a Radon barrier (pCi/cm^3)

LIST OF SYMBOLS AND ABBREVIATIONS cont.

Symbol

C_{34}	Radon concentration in the gas phase of soil before the application of a Radon barrier (pCi/cm ³)
$C(x)$	Radon concentration at x in the gas phase of a porous medium (pCi/cm ³)
C_o	Radon concentration in container (at/cm ³)
C_o'	Radon concentration in container (pCi/liter)
C_A	Radon concentration in basement of experimental building (pCi/cm ³)
C_c	Radon concentration in sampling container (pCi/liter)
C_f	Final Radon concentration (pCi/liter)
C_i	Initial Radon concentration (pCi/liter)
C_p	Radon plane source concentration
C_s	Radon concentration in gas phase of soil (at/cm ³)
C_t	Radon concentration in gas phase of tailings (pCi/liter)
$[C_{sol}]_{Ra}$	Radium concentration in solid phase of tailings (pCi/cm ³)
$[C_{sol}]_{Rn}$	Radon concentration in solid phase of tailings (pCi/cm ³)
CCl_4	Carbon tetrachloride
cm	Centimeter
cps	Centipoise
CS	Control Surface
CSU	Colorado State University
CV	Control volume
CNLM	Cumulative working level month
d	Diffusion coefficient (cm ² /s)
D	Decay rate of Radon atoms in the control volume (at/cm ³ -s)

LIST OF SYMBOLS AND ABBREVIATIONS cont.

Symbol

D_o	Diffusion coefficient of O_2-N_2 (cm^2/s)
D_{eff}	Bulk diffusion coefficient of the system N_2-O_2 in concrete (cm^2/s)
D_i	Conversion factor for the i^{th} gamma energy ($R\ hr^{-1}/photons\text{-}cm^2\text{-}s^{-1}$)
diam	Diameter
DMAPN	β - dimethylaminopropionitrile
DMT	N, N dimethyl-p-toluidine
E	emanation power of tailings
EPA	Environmental Protection Agency
F	measured flux out of concrete wall ($pCi/cm^2\text{-}s$)
F'	F/p
$F _x$	Flux of Radon atoms at location x ($at/cm^2\text{-}s$)
$f(x)$	Transcendental function in x
FRC	Federal Radiation Council
g	Gram
G_i	Frequency factor for emission of i^{th} gamma energy
G.S.D.	Geometric standard deviation
HCl	Hydrochloric acid
Hg	Element Mercury
hr	Hour
I_i^o	Initial activity of the i^{th} nuclide in a radioactive chain (atoms)
$I_i(t)$	Activity at time t of i^{th} nuclide in a radioactive chain (dis/time)
$I_i'(t)$	Activity at time t of i^{th} nuclide in a radioactive chain, expressed as a fraction of parent's activity

LIST OF SYMBOLS AND ABBREVIATIONS cont.

Symbol

ICRP	International Committee on Radiation Protection
in	Inch
JCAE	Joint Committee on Atomic Energy
K	Integration constant
k_o	Diffusion coefficient of Radon in air (cm^2/s)
k_c	Effective diffusion coefficient of Radon in concrete (cm^2/s)
k_c^*	k_c/porc
k_K	Effective diffusion coefficient in a porous medium for Knudsen diffusion (cm^2/s)
k_m	Effective diffusion coefficient in a porous medium for molecular diffusion (cm^2/s)
k_s	Effective diffusion coefficient of Radon in soil (cm^2/s)
k_t	Effective diffusion coefficient of Radon in tailings (cm^2/s)
k_{tr}	Effective diffusion coefficient in a porous medium for mixed diffusion
K-40	Isotope 40 of element Potassium
KFe	Potassium ferricyanide
kg	kilogram
l	Mean free path of diffusing substance in gas phase of porous medium
l_1, l_2	Distances travelled by gamma radiation in shields 1 and 2 (cm)
L	Linear dimension of porous medium
l_e	Effective distance travelled by an atom (molecule) to progress a linear distance L in a porous medium

LIST OF SYMBOLS AND ABBREVIATIONS cont.

Symbol

lb	Pound
m	Minute
M	Molecular weight of diffusing substance
MeV	Million electron-volt
ml	Milliliter
MMA	Methyl metacrylate
mR	MilliRoentgen
N_A, N_B	Molar fluxes of substances A and B
N_i	Initial population of i^{th} nuclide in a radioactive chain (atoms)
$N_i(t)$	Population at time t of i^{th} nuclide in a radioactive chain (atoms)
NBS	National Bureau of Standards
$\text{NH}_4(\text{OH})$	Ammonium hydroxide
NHPQ	Nelson, Haley, Patterson and Quirk (Engineering Consultants)
p	Porosity of medium
Pb-206	Isotope 206 of element lead
Pb-210	Isotope 210 of element lead
PC	Polymer-concrete
pCi	Pico-Curie (3.7×10^{-2} dis/s)
Po-210	Isotope 210 of element Polonium
Po-214	Isotope 214 of element Polonium
Po-218	Isotope 218 of element Polonium
porc	Porosity of concrete

LIST OF SYMBOLS AND ABBREVIATIONS cont.

Symbol

por _s	Porosity of soil
por _t	Porosity of tailings
ppm	Part per million
q	Square root of tortuosity factor
q ₁	$(\lambda \cdot \text{por}_t / k_t)^{1/2}$
q ₂	$(\lambda \cdot \text{por}_c / k_c)^{1/2}$
q ₃	$(\lambda \cdot \text{por}_s / k_s)^{1/2}$
Q.F.	Quality factor
r	$(\lambda \cdot \text{por}_c / k_c)^{1/2}$
$\bar{r}$	Average radius of the pores in a porous medium
R	Roentgen
[Ra]	Ra-226 content of the tailings (g/g)
rad	Radiation absorbed dose
rem	Radiological equivalent man
Ra-226	Isotope 226 of element Radium
[Rn]	Concentration of a Radon sample at the time of sampling (pCi/liter)
Rn-222	Isotope 222 of element Radon
RS	Experimental building. Reference building with slab on grade
s	Second
S'	Production rate of Radon atoms in the control volume (at/cm ³ -s)
S	Porosity of a medium
S _v	Photon source strength per unit volume (photons/cm ³ -s)

LIST OF SYMBOLS AND ABBREVIATIONS cont.

Symbol

$[S_v]_i$	Photon volume source strength of the i^{th} gamma energy (photons/cm ³ -s)
T	Thickness of concrete wall (cm)
T°	Temperature in degree Kelvin
t_1	Coordinate of tailings - concrete interface (cm)
t_2	Coordinate of concrete - basement atmosphere interface (cm)
t_i	Thickness of the i^{th} shield (cm)
T_a	Mean Air Residence time
T_s	Time lag between samples
TC	Concrete thickness
TiO ₂	Titanium oxide
TMPTMA	Trimethyl propane trimethacrylate
TS	Experimental building. Slab on grade with tailings
TT	Tailings thickness
U ₃ O ₈	Uranium oxide
USAEC	United States Atomic Energy Commission
USPHS	United States Public Health Service
V	Volume of sampling container
V_s	Volume of sampling apparatus
V_t	Volume of tailings
vs	Versus
w/c	Water/cement ratio by weight
WL	Working Level
WLM	Working Level Month

LIST OF SYMBOLS AND ABBREVIATIONS cont.

Symbol

X_A	Mole fraction of substance A
x_i	i^{th} estimate of the root of a transcendental equation
γ	$(k_c^*)^{1/2}$
Y	Overall yield of Radon scintillation chamber (count rate/alpha activity of sample)
Y_R	Radon yield of scintillation chamber (count rate/pCi of Radon)
yr	year
ZnS(Ag)	Silver activated Zinc sulfide
α_i	Correction factor of the linear attenuation coefficient
β	Constant related to the emanation power of a porous medium
λ	Radioactive decay constant of Rn-222
λ_i	Radioactive decay constant of the i^{th} nuclide in a radioactive chain
$\mu_{\text{Li}}, \mu_{\text{Zi}}$	Fictitious linear attenuation coefficient (cm^{-1})
μ_a	Linear attenuation coefficient of gamma radiation in air (cm^{-1})
μ_c	Linear attenuation coefficient of gamma radiation in concrete (cm^{-1})
μCi	Microcurie
μ_i	Linear attenuation coefficient of the i^{th} shield (cm^{-1})
μR	MicroRoentgen
μ_s	Linear attenuation coefficient of the material containing a radiation source (cm^{-1})
μ_t	Linear attenuation coefficient of gamma radiation in tailings (cm^{-1})
ρ	density (g/cm^3)

LIST OF SYMBOLS AND ABBREVIATIONS cont.

Symbol

ϕ_u	Uncollided flux (photons/cm ² -s)
ψ	Ratio of diffusion coefficients D_{eff}/D_o

SYMBOLS USED IN FORTRAN PROGRAMS*

Program KCANFLU

CS	Average Radon concentration in tailings gas phase (pCi/cm ³)
CC	Average Radon concentration in collecting device (pCi/cm ³)
F	Measured Radon flux (pCi/cm ² -s)
PORC	Concrete porosity
Y(I)	i th estimate of root for transcendental equation
AK	Effective diffusion coefficient of concrete (cm ² /s)

Programs BASFLUO and BASFLUS

RA	Radium content of tailings (g Ra-226/g tailings)
RO	Tailings density (g/cm ³)
F	Emanation power of tailings
AK1, AK2, AK3	Effective diffusion coefficients of Radon in tailings, concrete and soil respectively (cm ² /s)
PORS,PORT	Porosity of soil and tailings respectively
TH1,TH2	Thickness of tailings and concrete respectively (cm)
C _A	Radon concentration in basement of experimental buildings (pCi/cm ³)

*Symbols with similar meaning in subsequent programs are not repeated.

LIST OF SYMBOLS AND ABBREVIATIONS cont.

Symbol

N1, N2, N3 Number of computed values in Radon concentration and flux profiles in tailings, concrete and soil respectively

Program SLABCON

GAM(I) Energy of i^{th} gamma (Mev)

G(I) Frequency factor of i^{th} gamma energy (%)

TMU(I),
CMU(I),
AMU(I) Linear attenuation coefficients of i^{th} gamma energy in tailings, concrete and air respectively (cm^{-1})
(For air, 10^3cm^{-1})

A(I, 1),
A(I, 2) Weighing factors for experimental fit of buildup for i^{th} gamma energy

AL(I, 1),
AI(I, 2) Correction factors of the linear attenuation coefficients for the i^{th} gamma energy

CONV(I) Conversion factor for i^{th} gamma energy ($\text{R hr}^{-1} / \text{photon cm}^2\text{-s}$)

TT, TC, TA Thickness of tailings, concrete and air respectively (cm)

Program SLAPROT

D1, D2, D3 Relaxation distances for Radon concentrations in tailings, concrete and soil respectively (cm)

A14, B14,
A15, B15 Coefficients in Radon concentration profiles.
(see theory)

Program SLAPROC

A24, CA, B25 Coefficients in Radon concentration profiles.
(see theory)

1. NATURE OF THE PROBLEM

1.1 Introduction

Uranium mill tailings are a sand-like waste product which result from Uranium ore processing operations. Such operations have been conducted in nine western states of the United States, namely Colorado, Wyoming, South Dakota, New Mexico, Arizona, Utah, Washington, Oregon, and Texas. In some areas, a portion of the tailings have been used as a backfill material under and around buildings and as general fill for other construction projects, such as streets, sewer lines and sidewalks. The tailings contain minute quantities of Radium which decays into Radon and its daughters (Fig. 1.1). For the people living in the proximity of these tailings there is a possible health hazard due to an increase of the radiation exposure above the background level. There are two distinct types of exposures: first, the whole body gamma exposure is increased by the gammas which are emitted during the decay of the Radium daughters; second, there is an increased lung exposure to the alpha particles emitted by Radon and its daughters when these radionuclides are inhaled. In towns such as Grand Junction, Colorado, the problem assumes its full importance inside of certain structures where the use of tailings was extensive. In these structures the resulting increased levels of exposure reach values comparable to and sometimes higher than the recommended maximum permissible levels of exposure proposed by the International Committee on Radiation Protection (ICRP).

1.2 Discovery of the Problem - Initiation of the Present Study

1.2.1 Discovery of the problem. The problem created by the use of Uranium mill tailings as a construction material rose to such economic proportions for corrective measures that it raised the difficult question of the responsibility for causing the original problem and finally forced the intervention of the Federal Government. Furthermore there arose what were considered to be unjustified fears by the public because of inflammatory articles in the press. These combined situations prompted the Joint Committee on Atomic Energy (JCAE) to conduct public hearings before the Subcommittee on Raw Materials on October 28 and 29, 1971 [1.1]. In the statement of Mr. Robert Catlin, Assistant Director for Health Protection (Division of Operational Safety, AEC), we find [1.1 p. 43]: "The matter (increased radiation exposure due to Uranium mill tailings) was brought to the Commission's attention informally as a result of an outdoor survey of Radon and gamma radiation in the various Uranium mill towns. The USPHS (United States Public Health Service) was looking at the environmental situation which had been created in the vicinity of tailing piles ... At that time (1966) one of the surveyors went into a structure and noticed an elevated Radon reading within this structure. Following this ... in evaluating some Radon film badges, the State of Colorado personnel put some of these inside structures and noted that there were higher readings. They reported these back to their superiors." In the statement of Dr. Clarence E. Larson, Commissioner U.S.A.E.C. (United States Atomic Energy Commission) in the course of the same hearings, we find [1.1 p. 6]: "In July 1966, following reports of high Radon concentrations in a structure in Grand Junction (Colorado) by the Public Health Service, the Colorado Department of

Health personnel reported that tailings from the Climax* mill had been widely used as a construction fill in and around structure in that city ... An undetermined amount of tailings possibly from 200,000 to 300,000 tons had been distributed through the Grand Junction area from the Climax tailings pile between 1951 and 1966."

1.2.2 Initiation of this study. Environmental problems caused by Uranium mill tailings have been studied prior to the discovery of the current high indoor Radon concentrations in Grand Junction. An extensive bibliography of the subject can be found in the Appendix I of the JCAE hearings [1.1 p. 220]. However, as was noted in Dr. Larson's statement at the hearings [1.1 p. 6]: "The various analyses of risks posed by possible use of tailings included direct gamma radiation, inhalation of airborne tailings and the leaching of Radium in streams ... A thorough search of A.E.C. records discloses no early evidence that Radon from Uranium mill tailings would diffuse so readily into enclosed structures so as to constitute a source of risk. We find no references to the diffusion problem in Federal, State or industry correspondence or reports until the initial indoor Radon study by the Colorado Department of Health in 1966 and 1967 ..." Also as Dr. Larson indicates [1.1 p. 7]: "epidemiological study results on mortality of Uranium miners in relation to radiation exposure were first provided to the Federal Radiation Council in 1966." In Dr. Larson's opinion [1.1 p.7] these facts provide "the best explanation why no action was taken prior to 1966 to forestall the use of tailings for construction purposes."

*The Climax Uranium Co. which was a unit of the American Metal Climax Inc. has since become the AMAX Uranium Corporation.

The proposal for the present research project [1.5 p.3] was solicited from Colorado State University by the U.S.P.H.S. in the Spring of 1969 owing primarily to the previous experience of the principal investigators Dr. K.J. Schiager and Dr. H.G. Olson on Radon, its progeny in Uranium mines, and related subjects. The basic concept of the study is to test the effectiveness of various methods for reducing or eliminating the diffusion of Radon from Uranium mill tailings located under or around the foundations into buildings.

1.3 Origin and Radium Content of the Tailings

1.3.1 Origin of the tailings. Over the past few decades [1.1 p.5] about thirty-eight Uranium mills have been in operation in the Western States (12 in Colorado), producing an estimated 95 million tons of Uranium tailings. The function of Uranium mills is to extract Uranium (and sometimes associated elements, e.g. Vanadium at the AMAX mill in Grand Junction) in concentrated form from ores containing 2 to 3 kgs of Uranium oxide (U_3O_8) per ton of ore. These milling operations, after a series of extracting operations typified by Fig. 1.2 (Steps in Uranium and Vanadium ore processing) generate a residue (tailing) which contains small concentrations of radioactive materials, mostly Radium. The tailings come under two distinct types of product: one, the sand fraction, is a fine uniformly graded, brownish grey material. It is light in color when dry and dark when wet. The other fraction, the slime, is a fine powder when dry and has a purplish cast. It is discarded at a later stage in the refining process and has a higher Radium content than the sand.

1.3.2 Radium content of the tailings. Estimates of the Radium content of the tailings vary considerably. An assay by AMAX Uranium Co.

reports a content of 0.101 g Ra-226/1000 dry tons (111 pCi/g)* for the sand fraction and 0.592 g ²²⁶Ra/1000 dry tons (652 pCi/g) for the slime fraction [1.3]. Dr. Clarence E. Larson, a commissioner of the A.E.C. reports [1.1 p.5]: "The average Radium content of the tailings is in the order of 700 pCi/g, approximately the same as in feed ore." This last comment about the equivalence of Radium content for the feed ores and the tailings allows an estimate of an upper bound for the Radium content based on data for Radon flux out of ores. Dr. R.D. Evans [1.4 p.372] considers ordinary soil or rock to contain 2 μ Ci of Ra-226/cubic yard. For a density of the order of 3 g/cm³, this is equivalent to 1 pCi/g and these soils generate a Radon flux of about 1 μ Ci/day-square yard. In the Grants Uranium mines in New Mexico, the flux of Radon is 240 times greater than that from ordinary soils, and for sandstone Uranium ore faces the Radon flux is known to be even "several times greater" [1.4 p. 372]. Based on similar diffusion characteristics of the various soils mentioned, the Radon flux would be proportional to the Radium content and would approximately correspond to a Radium activity in the Uranium ore of 1000 pCi/g, i.e., about 1 g Ra-226/1000 dry tons. It is therefore reasonable to expect tailings to have a Radium content ranging from .1 to 1 g Ra-226/1000 dry tons of tailings.

*The Curie (Ci) is a unit of activity for radioactive nuclides; it corresponds to 3.7×10^{10} disintegrations per second. The picocurie (pCi) is 10^{12} times less, i.e., 3.7×10^{-2} dis/s.

1.3.3 Personal assay. As a part of this study, the physical characteristics of Uranium mill tailings, believed to be free of the slime fraction, were analyzed. A detailed description of these analyses is presented in Section 4. The Radium content of the tailings was found to be between 120 and 130 pCi/g tailings.

1.4 Tailings Usage

1.4.1 Spread in space and time. Even though the current attention has been focused on the misuse of tailings in the town of Grand Junction, Colorado, for the period between 1951 and 1966, the practice has not been limited to this sole period nor to this area. In August, 1969, [1.1 p 77], Union Carbide found high Radon levels in pre-1928 homes on company property in Uravan, Colorado. They moved the occupants from three homes and closed a bunkhouse. By examining old photographs of the town, it was concluded that these homes had been built over old Radium processing tailings (at that time there was no local effort to process Uranium ore; the desired product was Radium). Also there is evidence that tailings have been used at a sewage disposal plant in Utah and to fill a contractor's property in Salt Lake City [1.1 p 43]. As the surveys proceed, it is possible that uses of the tailings will be reported in all the locations where any of the 38 western Uranium mills are located.

1.4.2 Types of tailings uses in Grand Junction. The most thorough study on the variety of ways in which the Uranium mill tailings have been used has been conducted by the Colorado engineering firm Nelson, Haley, Patterson and Quirk in the town of Grand Junction [1.2 p. 356]. According to this report, the slime fraction of the tailings has no

particular value as a construction material since it retains water and use of it has probably been incidental because of accidental mixing in storage. The sand fraction, however, has some very desirable characteristics as a construction material: first, it compacts readily and makes good fill. In the particular case of the Grand Junction AMAX mill, its availability at no cost from a pile located in a central area of the city in a location where the clayish soil does not lend itself to easy compaction, resulted in its extensive usage. One of the most common uses of tailings was as a leveling course under concrete slabs. Many sidewalks, driveways, garage floors, footings and main floors of slab-on-grade houses were constructed by pouring concrete on a two to eight inch layer of tailings. Another common use was as backfill around foundation walls. The trench around the foundation wall was filled with tailings and usually covered with a layer of topsoil. In a relatively few cases, tailings were used as a general fill to raise the grade of a large area to the desired level. Water and sewer lines running underground from the house to the property line were sometimes laid on a bed of tailings in the bottom of the trench. Tailings were then used for backfill over the pipe with a layer of topsoil covering the backfill. Occasionally the tailings were used in mortar for masonry walls and fireplaces to lessen the sharpness effect of conventional mortar sand, but the substitution of tailings for more than 50 percent of the sand in the mortar made the mortar more difficult to work with and use in higher concentrations probably did not occur. Tailings were rarely used in concrete. They are found in concrete mostly in amateur construction projects. Some gardeners felt that tailings improved the tillability of the soil and mixed them into their flower and vegetable plots. Some

tailings were used as top dressing for lawns. AMAX Uranium Corp. estimates that 10 to 15,000 tons were also used by farmers who mixed the sand tailings to break up the shale lands to achieve better cultivation [1.1 p. 147]. Finally, considerable quantities of the tailings were also distributed as wind-borne dust, before the storage piles were stabilized by vegetation and water spraying.

1.4.3 Quantitative breakdown of tailings usage in Grand Junction.

An estimate of the quantitative breakdown of the uses of tailings was provided at the hearings by Mr. Anthony Mastrovich, Vice President of AMAX Uranium Corp. [1.1 p. 147]. According to this breakdown, 250,000 tons out of the 300,000 were used as a sub-base under roads, sidewalks and driveways and as packing around culverts, sewers and waterlines. The difference of 50,000 tons "probably is distributed around Grand Junction and its environs."

1.5 Mechanisms of Exposure to Radiations - Units

The mechanisms by which radiations reach the people who live in the proximity of the Uranium tailings are presented here. The various radiological units which will be often referred to are also described.

1.5.1 Gamma exposure. The Ra-226 present in the tailings is radioactive, with a half-life of 1602 years (for "half-life" see Appendix 1.1), and decays at a constant rate into Rn-222. The Radon itself is radioactive and decays into the next nuclide in the radioactive chain, Po-218. This radioactive decay from one nuclide into the next one goes on until the stable nuclide Pb-206 is reached. Figure 1.1 shows the nuclides of the chain which are of interest to us. It indicates that the decay of nuclides is often accompanied by the emission of gamma radiations. These radiations, which are of the same nature as

light, but of a higher frequency are highly penetrating. After being attenuated by the tailings and by the materials which exist between the tailings and the people, this type of radiation is still capable of penetrating the human biological system. In the body it may have direct or indirect destructive effects by disturbing the chemical organization of the cells. It should be mentioned that these phenomena occur constantly, even without the presence of radioactive tailings, because of the solar radiations and the radioactive nuclides present in any soil and in our body, an accumulation which is entitled "background." The effect of the tailings is merely to increase the exposure to such radiations above the normal background. Exposure rates to gamma radiations are measured in "Roentgens" (R). The Roentgen unit characterizes the intensity of the exposure by measuring the ionization which gamma radiations can induce in air. A person is said to be exposed to 1 Roentgen when he stands in a field of gamma radiation which induces 1 electrostatic unit of charge per cm^3 of air at standard pressure and temperature. The rate of exposure to gamma radiations will be stated in terms of Roentgens/unit of time. The Roentgen unit is only applicable to evaluate exposure to electromagnetic radiations like x-rays and gamma rays and is not used in the case of radiations involving particles like alphas and betas.

Gamma radiations can also be evaluated by the amount of energy they actually deposit in biological systems. This is measured by the unit called the "rad" (radiation absorbed dose). A dose of 1 rad means the absorption of 100 ergs of energy from ionizing radiation per gram of absorbing material [1.8]. In contrast to the Roentgen, the rad is

a unit used for any kind of radiation.. An exposure to 1 Roentgen of gamma rays will deposit approximately 1 rad in biological tissue so that exposure data expressed in Roentgens, for all practical purposes, can be converted directly to absorbed doses expressed in rads.

A third unit is used to evaluate the effect of ionizing radiations on biological systems. This last unit which, like the rad, applies to all kinds of radiations, is called the "rem." Since the various types of radiation deposit their energy by different mechanisms with very different resulting biological effects a "quality factor" (Q.F.) was devised. By multiplying the actual dose received (in rads) by the quality factor (Q.F.) one obtains the "dose equivalent" in rems:

$$\text{rem} = \text{rads} \times \text{Q.F.}$$

The quality factor for gamma radiation is the lowest one and is equal to 1. Absorbed doses expressed in rads can thus also be expressed in rems.

1.5.2 Radon and Radon progeny exposure. As mentioned earlier, the Ra-226 present in the tailings decays with a half-life of 1602 years to Rn-222 and constitutes a constant source of Radon for a period of time much larger than the lifetime of the constructions in which tailings were used. Radon is the heaviest of the inert elements chemically classified in the last column of the Mendeljev table which includes Helium, Neon, Argon and Krypton; it is an inert monoatomic gas which means that its affinity for other atoms is very low. As a result, the mobility of Radon atoms is very large and they diffuse readily through most materials. After escaping from the tailings and diffusing through the materials which cover or are adjacent to the tailings, the Radon becomes airborne and can be inhaled by the people;

yet, despite its activity as an alpha emitter, Rn-222 itself presents little hazard. Its inert characteristics result in a low retention in the lungs and its half-life of 3.82 days is long compared to the respiration cycle, so that a negligible fraction of it decays by alpha emission while present in the lungs. The short-lived Radon daughters which appear in the atmosphere, as the airborne Radon decays, present a greater health risk. When airborne Radon decays into its daughters, they attach to the particulates in the air and show a greater retention in the lungs than the inert Radon. They range from the Po-218 to the Po-214. Pb-210 is not included in the isotopes of concern in the lungs because its long half-life (21 years) ensures that the great majority of the Pb-210 atoms will be eliminated from the respiratory system before they decay by emission of gamma and beta radiations. The harmful effects, if any, are therefore to be expected from the short-lived Radon progeny. By analogy with the gamma radiations, it should be mentioned that exposure to Radon daughters occurs even without the presence of radioactive tailings. Normal soils, as previously mentioned, contain a certain amount of Radium. The Radon they release is responsible for a background level in Radon progeny. The effect of the Uranium tailings is to increase the concentration of the Radon daughters above this background level. This effect is only noticeable indoors. Outdoors, the increase in the Radon progeny, concentration due to the tailings is negligible because of the rapid dilution taking place in the atmosphere.

Exposure to Radon daughters is measured in "Working Level" (WL) units. This unit is based on the observation that two of the Radon daughters, the alpha emitters Po-218 and Po-214, are responsible for

most of the dose absorbed by the lung cells. The exposure to Radon, more accurately to Radon progeny, is therefore measured by the concentration in the air of short lived Radon daughters. This concentration is then converted to the potential alpha radiation which will ultimately be emitted from a mixture of these nuclides. One WL is defined as any combination of short lived Radon daughters per liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha energy by ultimate decay to Pb-210. The value of 1.3×10^5 MeV of alpha energy is that which results from the decay of Radon progeny in secular equilibrium with one hundred pCi of Radon. A radioactive chain is in secular equilibrium with the parent when all the daughters are being formed at the rate at which they are decaying. In the case of continuous supply of the chain parent, the total activity of each radionuclide remains constant and each nuclide in the chain shows the same activity as the parent of the chain (See Appendix 1.2).

The cumulative exposure to Radon progeny is measured in Working Level Month (WLM), i.e., the exposure of a person residing during a working month (170 hours) in a concentration of 1 WL or in Cumulative Working Level Month (CWLM) which is computed by multiplying the period of exposure, expressed in month, by the Radon progeny concentration expressed in Working Levels. Fig. 1.3 [1.1 p. 208] presents a visualization of the mechanisms by which people are exposed to radiations indoors. These exposures exist in every structure. The effect of the tailings is to increase the level of exposure.

1.6 Exposures Allowed by Guidelines. Actual Exposure Increase Above Background in Grand Junction.

All the data relative to the increased exposure due to the tailings are derived from the city of Grand Junction. To date it is the only location where thorough investigations have been conducted.

1.6.1 Gamma radiation.

1.6.1.1 Problems in assessing exposures. This type of exposure is the easiest to assess. Provided an accurate calibration of the field instrumentation has been performed, the constancy of the gamma field created by the tailings allows rapid and accurate measurements.

1.6.1.2 Guidelines for gamma exposures. Publication No. 9 of the International Committee on Radiation Protection (ICRP) [1.6] and Report No. 1 of the Federal Radiation Council (FRC) [1.8] both recommend that the maximum total annual whole body exposure of the general public to external gamma radiation be limited to 500 mR/year. This excludes background radiations and the exposures incurred under medical treatment. The gamma radiation background in Grand Junction has been measured by the Colorado State University personnel using a pressurized ionization chamber; backgrounds around 0.014 mR/hr ($\approx$ 123 mR/year) have been measured ([1.5 p. 2] and [1.1 p. 53]). For comparison, an AEC publication on radiation in the environment [1.7] lists 100 mrem/year as an average total dose rate due to external and internal irradiation from natural sources in a normal area. In the United States, it varies from 80 to 200 mR/yr [1.11].

On July 27, 1970, Dr. Jesse L. Steinfeld, Surgeon General of U.S.P.H.S. responded to the Colorado Department of Health request for guidelines about the Grand Junction problem, by recommending that

actions be taken in the case of high radiation levels in dwellings constructed on or with Uranium mill tailings. For the external gamma irradiation the recommendations were stated as follows in terms of graded action levels:

Exposures in Addition to Natural Background:

([1.9] and [1.1, p. 214])

<u>External Gamma Radiation</u>	<u>Recommendation</u>
1. Less than 0.05 mR/hr	No action indicated
2. From 0.05 to 0.1 mR/hr	Remedial action may be suggested
3. Greater than 0.1 mR/hr	Remedial action indicated

In the case of a 24 hr/day occupancy, levels 1 and 3 result respectively in exposures of 438 mR/yr and 876 mR/yr. There has been some concern about the indecision area left by these guidelines in a range extending by nearly a factor of 2 above the ICRP maximum. A justification of this approach can be found in the explanatory note to the guidelines [1.9]: "This graded system of action is proposed to allow latitude in the middle ranges for the judgment of in site investigators," Also [1.9]: "The upper limit of these ranges exceed the strictly applied recommendations of the FRC and ICRP for exposures of an individual member of the public. However, this extension seems justified in situations in which unforeseen exposures have occurred, since as stated by ICRP, (5)*: In general it will be appropriate to institute countermeasures only where their social cost and risk will be less than

*[1.6] in our references.

those resulting from the exposure. It is further stated by ICRP (5)* that very low levels of risk are implied in the dose limits for members of the public and that it is likely to be of minor consequence to their health if the dose limit is marginally or even substantially exceeded." These remarks of the Surgeon General take all their significance if one considers that the limits set by the ICRP for exposures of people occupied in activities involving irradiation (occupational exposures) are established with the absolute safety of the people in mind; yet they exceed the limit for the general public by a factor of 10 (limits for occupational exposures have been set at 5000 mR/year).

1.6.1.3 Gamma exposure in Grand Junction. As of October 1971, the gamma survey conducted by the Environmental Protection Agency (EPA) in Grand Junction provided data for 1,037 structures out of the 5,000 which were identified as possibly having tailings used in their construction on the basis of an uncorrected gamma reading of 20 μ R/hr. These data include 777 residences, 194 businesses, 4 schools, and 42 buildings owned by the public such as churches, a post office, airport buildings, etc. The results are shown in Table 1.1. Except for the schools, which numerically constitute the least important group, the great majority of the structures with tailings do not exceed the ICRP limit for exposure of the public to gamma radiation. This consideration is of importance in analyzing potential remedial solutions.

1.6.2 Radon daughters

1.6.2.1 Problems in assessing exposures. The annual average exposure of people to the Radon progeny is much more difficult to assess than the gamma exposure. The reason is that a structure built

over a given amount of radioactive tailings will show Radon and Radon daughter concentrations which vary greatly in time. These variations depend on well known, but hardly measurable factors, like the degree of ventilation of the house, but also on hypothetical factors like the variation in atmospheric pressure which probably influences the rate of effusion of Radon out of the tailings into the structures. Several witnesses, present at the JCAE hearings, reported the difficulties encountered in determining an accurate yearly average of Radon concentration in a given location. Mr. Robert Catlin, for example stated [1.1 p. 22]: "The range of variation in regard to Radon levels in houses varies throughout the year by perhaps a factor of 10 or 100, one sample to the next. This is because of differences in climate, temperature, degree of ventilation, whether the heating plant is on or not, and so forth." He also stated [1.1 p. 47]: "In the summertime, the Radon levels might be quite low because the house would be ventilated well. In the wintertime, the Radon levels might rise except if there were electrostatic precipitators or other air cleaning systems, these might take some of the Radon daughters out." Mr. David D. Dominick, reporting as the Assistant Administrator of the EPA, stated during the same hearings [1.1 p. 74]: "The Radon concentration in a structure built with tailings does not depend only on the amount of tailings present. The location of the tailings, the type of construction, the amount of outside ventilation, and the patterns of air flow within a structure all affect Radon daughters concentrations. In addition, climatic conditions, principally barometric pressure and temperature can affect the levels as well as secondarily affecting the ventilation and airflow patterns within the structures."

To add to the difficulty, measured concentrations of Radon gas do not necessarily provide the concentration of Radon daughters on which the computation of Working Levels is based. The equality in activity of Radon with its daughters is only reached in an equilibrium situation in which none of the nuclides in the decay chain is selectively removed. On site, however, the difference in the chemical properties between Radon and its daughters does lead to a selective removal of the Radon daughters by plating out on surfaces and in ventilation devices. This in turn results in variable discrepancies between the activity of Radon and the activity of its progeny in a room. This variability is confirmed by the data obtained in the experimental structures built on the campus of Colorado State University for the study of possible remedial actions to the Grand Junction problem. These windowless buildings (20' x 10') were standardized with respect to the air mean residence time. Radon as well as Radon progeny concentrations were monitored over a period of seven months. The data are given in the first progress report of the project [1.5 p. 80] and read: "The geometric standard deviations of the measurements of concentrations in the experimental modules were approximately 1.5 for Radon progeny and 3 for Radon. For these conditions of relatively stagnant indoor atmospheres (mean air residence time of approximately 3 hours), the percentages of individual measurements expected to fall within factors of 2, 3, or 5 of the long-term mean concentrations are as follows:

	Percentage of individual measurements expected between Mean/F and Mean x F		
	F = 2	3	5
Radon progeny (G.S.D. = 1.5)	82%	95%	99%
Radon (G.S.D. = 3)	50%	68%	90%

The report continues: "In occupied buildings with mechanical ventilation and people moving in and out and between rooms etc., the expected variability would be much greater than that observed in the experimental modules. Also, under comparable conditions of measurement, the variability of Radon progeny concentrations would be expected to be substantially greater than that of Radon gas, as a result of removal by furnace filters, plate-out in furnace duct work and on other surfaces etc. ..."

As a result of this variability it was decided that average Working Levels in a given location would have to be based on air samples collected throughout the year with a minimum of six one-week-air samples at intervals spaced over a whole year.

1.6.2.2 Guidelines for Radon progeny exposure. Prior to the discovery of the increased indoor Radon concentration in structures built over Uranium mill tailings, there existed no specific guidelines for the exposure of the general public to Radon progeny. However if one follows the policy recommended by ICRP [1.6] that the annual maximum exposure of individual members of the public to any type of radiation be kept to one tenth of the corresponding maximum annual occupational exposure, one should use 0.4 WLM/year. This is one tenth of the 4 WLM/year limit recommended by the FRC [1.10] as a maximum occupational exposure to be implemented on January 1, 1971.

In the letter referenced before [1.9], the Surgeon General of the United States also made recommendations relative to the exposure to Radon progeny. These recommendations are:

Exposures in Addition to Natural Background:

<u>Level</u>	<u>Recommendation</u>
1. Less than 0.01 WL	No action indicated
2. From .01 to 0.05 WL	Remedial action may be suggested
3. Greater than 0.05 WL	Remedial action indicated

In the case of a 24 hrs/day continuous exposure, levels 1 and 3 amount to 0.52 WLM/year and 2.6 WLM/year.* The Radon progeny background in Grand Junction has been measured by the Colorado Department of Health and was found to be .004 WL. This corresponds to 0.2 WLM/year in the case of a 24 hrs/day exposure. The lower level of the Surgeon General's guidelines is therefore about 30% higher than the maximum exposure recommended by the ten percent ICRP rule and the higher level is about 5 times higher than the same ICRP guideline. By comparison to the position taken with respect to the gamma exposure, not only is the "no action" level higher than the ICRP guideline but the indecision area is defined by a factor of 5 while it involved a factor of 2 for the gamma exposure. A justification of these facts is found in the deposition of the Surgeon General himself at the JCAE hearings [1.1 p. 58]: "The reason for this relates to the reliability of the measurement. It is much more difficult to measure Radon daughter products. As you know, the Radon daughter products in these houses vary with the time

*The WLM unit is based on an exposure of 170 hrs/month or $12 \times 170 = 2040$ hrs/year while there are 8766.1 hrs in a year; i.e., $8766.1/2040 = 4.39$ times more.

of the year, with whether or not the windows are open, with whether or not the heating system is working and even the instrument for measurement of Radon daughter product is not as reliable as is the gamma instrument. We felt we needed a wider range of latitude for the exercise of judgment in the case of Radon daughter products." The other justifications given for the gamma exposure guidelines also apply in the case of Radon progeny.

1.6.2.3 Radon progeny exposure in Grand Junction. The situation in Grand Junction with respect to the concentration of Radon progeny has to be evaluated from less abundant data than the exposure to gamma radiation because of the lengthy measurements required to evaluate it. Structures with tailings are presently being surveyed for Radon progeny. As of October 1971, 22 structures had been sampled 6 times and 27 structures 5 times with one week-long samplings. Besides these 49 almost complete results, 14 houses without tailings had been surveyed. The distribution of the Radon progeny concentration is shown in Table 1.2 [1.1 p. 217]. None of the control locations were found to reach the Surgeon General's level 1, but 87%* of the houses with tailings show concentrations equal to or above the .01 WL concentration. The large proportion of houses (87%) with Working Levels above the set guidelines is in sharp contrast with the one observed for structures with a gamma level above the recommended level (1.7% for residences to 41.6% for schools). This situation is also important in considering remedial actions.

*This number was later found to be an underestimation. The readings of the integrating air samplers used to measure average Working Levels in the houses were found to require a correction factor of 2 to account for the Radon progeny plate-out which was taking place in the inlet of the samplers [1.12].

2. MOTIVATION AND ORGANIZATION OF THE STUDY

2.1 Primary Motivation of the Study.

As has been mentioned previously, tailings which result in outdoor exposures are less critical because the Radon progeny appearing outdoors is rapidly diluted in the atmosphere. Also, the exposure to the gamma radiation is often of a transitory nature as opposed to the continual exposure of the people living in a structure. The most critical situation has therefore been created indoors because of the high Radon progeny concentrations which have developed.

The distribution of structures with tailings versus the level of exposure to each type of irradiation, gamma (Table 1.1) and Radon progeny (Table 1.2) clearly shows that in the great majority of the cases the levels of exposure above permissible limits arise from the Radon progeny concentration whereas gamma exposure exceeds the permissible levels only in a few percent of the cases. The problem would therefore be corrected in the majority of the contaminated houses if a method was devised to reduce the concentration of Radon progeny in a structure. One approach to the problem consists of eliminating the source of Radon emanations and proceed to the removal of all the Uranium mines tailings from the structures foundations. As will be shown in the following section, the economic aspect of the removal solution becomes critical when tailings used in structures are of difficult access. This observation prompted the present work which analyzes a method of reducing Radon infiltration into structures by means short of tailings removal.

The cases of tailings use considered in the present work are the ones in which tailings were used under or around structures with a resulting gamma radiation below the safe level, but in which the Radon

progeny concentration is too high. These cases constitute a majority. For the houses where the gamma exposure constitutes the problem, installation of radiation shields or removal of the tailings from under and around the structures are the only options left.

2.2 Supplemental motivation.

In addition to the need for a remedial solution to the Grand Junction type problems, the need for a product which would stop the diffusion of Radon inside structures has been expressed [2.1] by the Health Physics staffs responsible for monitoring the airborne alpha activity in buildings with massive concrete walls. The lack of discrimination of the gross alpha monitors, installed to detect any leak of alpha emitting nuclides handled in facilities such as nuclear reactors and industries handling Plutonium, generates alarm signals in instances of low ventilation because of the indoor accumulation of Radon born from the decay of the Radium present in the massive concrete walls. A coating capable of stopping the natural Radon influx in these facilities would be helpful in allowing one to set the monitoring instrumentation at low alarm levels, associated with early detection of leaks, without continuously risking spurious triggers due to temporary increased levels in Radon progeny.

2.3 Extent of the problem and evaluation of remedial actions.

2.3.1 Extent of the problem created by the use of tailings as a construction material. In exercising a choice between several effective remedial solutions, one should certainly consider the cost of the solution. This becomes very apparent upon consideration of the possible extent of the problem. The town of Grand Junction is the only one for which enough data are available to permit a sound extrapolation. In

the attachment to the statement of Mr. Robert Siek, at the time Assistant Director of the Occupational and Radiological Health Division, Colorado Department of health, we find [1.1 p. 218]: "An estimate of 18,000 habitable structures in the Grand Junction area can be obtained from the 1970 census data for Colorado. If the gamma screening trend observed continues (Table 4)*, about 20 percent of the structures surveyed, or about 3,600 will have tailings located against or under the building. Assuming that the data of Table 3** are representative, approximately 3,100 structures can be expected to have average Radon daughter concentration of .014 WL or higher." If a similar situation is found in the other communities possibly involved, it is apparent that the unit cost of the remedial solution will be multiplied by a factor of such a magnitude that it becomes mandatory that the adopted remedial solution not only be effective but also be the least expensive.

2.3.2 Solution by tailings removal. Cost estimate. The most efficient solution with respect to reduction of exposure consists in removing the tailings and substituting them with soil having a normal Radium content. Yet, as we can find in the deposition of Dr. Larson at the hearings [1.1 p. 16]:

"A logical question frequently raised by individuals is 'in view of the uncertainties in estimating possible additional risks to health and the difficulty in decision-making, why not just remove all tailings and be done with it?' The following considerations militate against this approach:

*Table 2.1

**Table 1.2

1) A strong official recommendation to remove all tailings, however insignificant the associated radiation exposure or impact of remedial action, would place an unwarranted stigma on a large number of structures where the health risk is very slight or nonexistent. This stigma could extend to structures in other states if similar exposure conditions prevailed, even though tailings were not present.

2) The general public might be convinced that action was necessary wherever there were variances in exposure from natural sources. As stated previously, about 20 million persons are exposed to natural radiation levels in excess of mean levels in Grand Junction.

3) The cost of removal of all tailings under or against structures in Grand Junction alone would be large, possibly in the ten's of millions of dollars. In our opinion, the expenditure of this much money is warranted only if the incremental reduction in risk is significant compared to the cost of that reduction. As noted in FRC Report No. 5, 'Proposed protective actions must be weighed against their total impact. ... As the projected doses become less, the value of protective actions becomes correspondingly less.' "

Nevertheless, because the removal solution will most probably be applied in the cases of high gamma readings and in the cases of easy access to the tailings, and also because one needed an upper bound on the cost of the other potential solutions, the AEC contracted the Colorado engineering firm NHPQ to study the feasibility and the cost of removing the tailings from under or against residences [1.2]. For this study 240 residences were chosen out of a list of homes which showed evidence of tailings on the basis of preliminary on-site gamma radiation surveys. The 240 homes were then selected randomly so as to provide a representative sample of various types of constructions and tailings use. The condensed cost estimate data are the following:

"Of these (240 residences), 25 were not estimated because the presence of tailings could not be established. Cost estimates ranged from \$43 for a category 311 residence (crawl-space, some masonry, tailings under or against the structure) where some small deposits of tailings were

located on the surface of the ground near the house to \$15,000 for a category 123 residence (basement, non-masonry, tailings under or away from structure) where tailings were used extensively under various slabs. The average estimated cost for tailings removal from the residences was \$3,220 but the average estimated costs in individual categories ranged from \$1,400 to \$6,335.

The most frequently encountered situation (12% of total) involved a house built on a crawl space with an attached garage. The garage slab was poured on a layer of tailings. There was usually a patio slab also poured on tailings. The average estimated cost of tailings removal for this combination was \$2,460 with cost estimates ranging from \$472 to \$6,649."

Since several radiation protection methods will probably be used on different parts of the same structure, a better base of comparison for the cost of different remedial solutions can be obtained from the unit cost estimate data contained at the end of the same report [1.2]. These data were derived from the detailed estimates of the residences in the same study. The costs include related items such as removing furniture, protecting adjacent equipment and restoration. The reported costs can be found in Table 2.2.

If other efficient remedial methods are considered they would have to cost significantly less than the prices cited in Table 2.2.

2.3.3 Solution by ventilation. High Radon progeny concentrations is a problem which has been encountered in Uranium mines before. The solution adopted in the case of enclosed and well channeled mine galleries is to reduce the Radon progeny concentration by an appropriate ventilation system. This ventilation either replenishes the mine atmosphere by fresh air brought from ground level or it recycles it in a closed circuit by passing it through filters which collect some of the Radon daughters. The recycled air is then said to be "young" in the sense that the selective removal of Radon daughters keeps them from

reaching an equilibrium activity equal to the high Radon activity found in the mine. This practice results in lowering the level of exposure to Radon progeny in spite of a high Radon concentration.

The ventilation solution was rejected in the case of residential buildings mainly because it requires a constant maintenance of the ventilation equipment which would have to run on a 24 hrs/day basis. Not only would this solution lead to some inefficiency due to the negligence of certain homeowners, but it would also reduce the value of a residence. Finally, it appears less feasible to establish a thorough ventilation system in a multiple-story, multiple-entry residence than in a system of mine galleries.

2.3.4 The inner Radon barrier solution. As an alternative to the removal of the tailings, it was observed that, in the houses where the problem consisted only of a high Radon progeny concentration (the majority of the cases), the implementation of means to stop the Radon from seeping in the structures might be economically more advantageous, while as efficient. If successful, the method would not necessarily be applied in every case but only in those locations where the difficult access to the tailings renders the removal operation much more expensive than the application of Radon barriers. These instances include the presence of tailings under foundation slabs of houses with basement, crawl-space or slab-on-grade, tailings under garage slabs and tailings against support foundation walls of difficult access because of their location with respect to the superstructures (Fig. 2.1).

Two procedures can be followed in applying Radon barriers. The first one consists of drilling the slabs to be sealed and injecting a substance which either forms a sealing coat between the tailings and the outer surface of the slab, or conglomerates the tailings in a mass impervious to Radon, or perhaps achieves both effects. A few such products have been tested at Colorado State University. The other procedure, which requires less perturbation of the structure foundations, consists of applying a coating impervious to Radon on the indoor face of the walls to be sealed. This coating would suppress or at least reduce the Radon influx either by blocking it or by retarding its diffusion so much that most of the Radon has decayed into its daughters before it can emerge inside of the structure. The daughters which do not possess the inert characteristics of Radon could not diffuse across the barrier. This second method is the one analyzed in the present work.

2.4 Organization of the Study. The development of a testing facility and the results obtained with various products applied as Radon sealants are presented in Chapter 7.

Prior to conducting experiments in order to discover a material with good wearing properties which would be partially or totally impervious to Radon, the feasibility of the inner coating approach required that the following problem be quantitatively answered: as the Radon which diffuses from the tailings is blocked by an impervious coating, its concentration in the wall or floor slab increases above the value which existed while it was free to diffuse out of the slab. With the Radon concentration, the concentration of progeny also increases.

The result is an increased gamma exposure due to the decay gamma radiations of the Radon progeny trapped in the slab. This problem, which would also exist to a lesser extent in the case of an outer Radon barrier, required a quantitative evaluation of the increase in gamma exposure. The solution to this problem involved two aspects. First, the diffusion of Radon had to be quantitatively described in the concrete walls of a structure as well as around it, in order to evaluate the consequences of a sealant application in terms of Radon concentration buildup. This was accomplished by the establishment of reliable instrumentation for the evaluation of Radon concentrations (Chapter 3), by the measure or the study of the physical properties of Uranium mill tailings and concrete (Chapter 4) and by the application of a linear diffusion model to a multilayered system of porous media consisting of soil, tailings and concrete (Chapter 5). Next, the diffusion of Radon being appropriately described, a calculation of the increase in gamma exposure resulting from a sealant application was performed for various cases with emphasis on the worst possible cases (Chapter 6). An experimental facility was then devised to test the permeability of various coatings to Radon (Chapter 7) and conclusions were drawn on the remedial solution analyzed in this work (Chapter 8).

3. INSTRUMENTATION

The potential hazards resulting from the exposure to high Radon concentrations arise from the short lived Radon progeny. Nevertheless, our efforts, in this study, were directed toward the measure of Radon concentrations because it is the influx of Radon and its rate of leakage which ultimately control the amount of progeny in a building. Radon concentrations were determined by means of Radon scintillation chambers. The evaluation of the Radon content of a sample, by this technique, relies on the alpha activity of Radon as well as on the activity of two short lived daughters (Po-214 and Po-218) after the sample has been maintained in a sealed scintillation chamber for a predetermined period of time.

3.1 Radioactive Decay Chains

Because of the extensive use which is made of the Radon progeny in determining the actual concentration of the parent in a sample, it is often required to evaluate the distribution of relative activities of the various Radon daughters in a sample, after a specified period of time. This is provided by the solution to the equations which describe the time dependence of the nuclides' activities in a radioactive chain. The solution to this problem was presented by H. Bateman in 1910 [3.1] in the form shown in Fig. 3.1. The relations presented in Fig. 3.1 cover the case of a chain consisting of four nuclides. The rate at which the expressions grow in complexity with the number of nuclides considered revealed this formulation to be of little practical use in the cases encountered in our work since we considered up to ten nuclides. A general solution which would

easily lend itself to the operations of a digital computer was developed in the following manner:

Consider a sealed enclosure containing an initial population of n nuclides ranging from a parent 1 to the last nuclide in the series, n , which is stable. The initial condition is given by

$$N_i(0) = N_i^0 \quad (i = 1, \dots, n) \quad (3.1)$$

and the equations which describe the time evolution of the n populations are:

$$N_1(t) = N_1^0 e^{-\lambda_1 t} \quad (3.2)$$

$$\frac{dN_i(t)}{dt} = \lambda_{i-1} N_{i-1}(t) - \lambda_i N_i(t) \quad (3.3)$$

where λ_i are the respective decay constants of the radioactive nuclides, with $\lambda_n = 0$ since it is stable.

The dependence of the i^{th} population on all the ones which precede it in the chain, suggests a solution of the type

$$N_i(t) = \sum_{j=1}^i A_{ij} e^{-\lambda_j t} \quad (3.4)$$

The problem reduces then to find a general formulation for the A_{ij} 's. Substituting (3.4) into (3.3), we obtain

$$\frac{dN_i(t)}{dt} + \lambda_i N_i(t) = \lambda_{i-1} \sum_{j=1}^{i-1} A_{i-1,j} e^{-\lambda_j t} \quad (3.5)$$

The integration of (3.5) can be readily achieved by multiplying Eq. (3.5) by the integrating factor $e^{\lambda_i t}$:

$$e^{\lambda_i t} \frac{dN_i(t)}{dt} + \lambda_i N_i(t) e^{\lambda_i t} = \lambda_{i-1} \sum_{j=1}^{i-1} A_{i-1,j} e^{(\lambda_i - \lambda_j)t} \quad (3.6)$$

which is equivalent to

$$\frac{d}{dt} [e^{\lambda_i t} N_i(t)] = \lambda_{i-1} \sum_{j=1}^{i-1} A_{i-1,j} e^{(\lambda_i - \lambda_j)t} \quad (3.7)$$

By calling K the integration constant, (3.7) integrates to

$$e^{\lambda_i t} N_i(t) = \lambda_{i-1} \sum_{j=1}^{i-1} \frac{A_{i-1,j}}{\lambda_i - \lambda_j} e^{(\lambda_i - \lambda_j)t} + K \quad (3.8)$$

A solution for any population $N_i(t)$ is obtained by multiplying (3.8) by $e^{-\lambda_i t}$:

$$N_i(t) = \lambda_{i-1} \sum_{j=1}^{i-1} \frac{A_{i-1,j}}{\lambda_i - \lambda_j} e^{-\lambda_j t} + K e^{-\lambda_i t} \quad (3.9)$$

where the integration constant K remains to be evaluated. Substitution of the initial conditions (3.1) into (3.9) provides

$$K = \frac{N_i^0}{\lambda_{i-1}} - \sum_{j=1}^{i-1} \frac{A_{i-1,j}}{\lambda_i - \lambda_j} \quad (3.10)$$

Substitution of (3.10) into (3.9) leads to the final solution:

$$N_i(t) = \sum_{j=1}^{i-1} \frac{\lambda_{i-1} A_{i-1,j}}{\lambda_i - \lambda_j} e^{-\lambda_j t} + [N_i^0 - \sum_{j=1}^{i-1} \frac{\lambda_{i-1} A_{i-1,j}}{\lambda_i - \lambda_j}] e^{-\lambda_i t} \quad (3.11)$$

The recurrence aspect of Eq. (3.11) is best seen when written in the form

$$N_i(t) = \sum_{j=1}^i A_{ij}^* e^{-\lambda_j t} \quad (3.12)$$

where

$$A_{ij}^* = \frac{\lambda_{i-1}}{\lambda_i - \lambda_j} A_{i-1,j}^* \quad (j = 1, \dots, i-1) \quad (3.13)$$

$$A_{ii}^* = N_i^0 - \sum_{j=1}^{i-1} A_{ij}^* \quad (j = i) \quad (3.14)$$

The system (3.12), (3.13) and (3.14) provides a solution for the time evolution of each nuclide in the chain, in terms of the coefficients describing the preceeding nuclide in the chain. The formulation is complete since Eq. (3.2) provides the first coefficient $A_{11} = N_1^0$ and it lends itself easily to computation by digital computer through the use of subscripted variables.

Very often, the required solutions involve the activities $I_i(t)$ of the various nuclides rather than their respective populations. The required solution is obtained through multiplication of (3.12) by λ_i . This leads to the following solution:

$$I_i(t) = \sum_{j=1}^{i-1} B_{ij}^* e^{-\lambda_j t} \quad (3.15)$$

where

$$B_{ij}^* = \frac{\lambda_i}{\lambda_i - \lambda_j} B_{i-1,j}^* \quad (j = 1, \dots, i-1) \quad (3.16)$$

$$B_{ii}^* = I_i^0 - \sum_{j=1}^{i-1} B_{ij}^* \quad (3.17)$$

where I_i^0 = initial activity of the i^{th} nuclide. Finally, it also commonly required to obtain the activity of the daughters as a fraction of the parent's activity. This is achieved through division of Eq. (3.15) by the activity of the parent, $I_1^0 e^{-\lambda_1 t}$. The solution is then given by the system (3.16), (3.17), (3.19) and

$$I_i'(t) = \sum_{j=1}^i \frac{B_{ij} e^{(\lambda_1 - \lambda_j)t}}{I_1^0} \quad (3.18)$$

where $I_i'(t)$ = activity of the i^{th} nuclide as a fraction of the chain parent's activity.

Appendix 3.1 reproduces a FORTRAN program, "DEC SER," which represents the above development.

3.2 Radon Scintillation Chambers

3.2.1 Principle of operation and fabrication. Radon scintillation chambers use the characteristic alpha particles emitted by Radon and two of its short lived daughters (Po-214 and Po-218) to determine the Radon concentration of an air sample. The technique, proposed by Van Dilla and Taysum [3.5], was used in this work under the modified form proposed by Lucas [3.2]. The air sample is sealed in a container which has part of its surface replaced by a glass window, while the inner surfaces of the remaining walls are coated with Silver activated Zinc sulfide (ZnS(Ag)). The latter product exhibits the long known property (Rutherford) of emitting photons in the visible range when struck by alpha particles, photons or gamma rays. The lower intensity of the light pulses triggered by gamma rays allows them to be discriminated and, if the counting is performed in the dark, the photoactivity of the container is directly proportional to the total alpha activity inside the container. The Radon activity of the air sample can then be deduced if the relative activities of the other alpha emitting daughters is known. This last information is obtained by an analysis of the time evolution of a Radon sample with respect to the buildup in activity of its daughters.

Metal Radon scintillation chambers are commercially available at a minimum price of \$220. Since the project made extensive use of scintillation chambers (40 were used simultaneously), an attempt was made to avoid the high capital cost and fabricate them in the Department

of Radiology and Radiation Biology under the supervision of Dr. K. J. Schiager and his doctoral student D. R. Alexander. Nalgene pharmaceutical graduates (250 ml) were used as containers and sawed at a level which allowed the fitting of a 2" diameter quartz window. The ZnS(Ag) coating was obtained by spraying a mixture of ZnS(Ag), quick drying varnish (for bonding), and acetone (for dilution) onto the walls of the flask. A sufficient amount of the mixture was sprayed to attain a ZnS(Ag) density thickness larger than the required 20 mg/cm^2 for optimum yield [3.2]. The type of ZnS(Ag) powder was chosen so that its bimodal photoresponse would be almost totally covered by the response curve of the photomultiplier tube (3000 to $6500 \text{ \AA}$). The quartz window was verified to have a good and constant transmission characteristic over the range 3000 to $6500 \text{ \AA}$.

The use of self-made scintillation chambers necessitated calibrations and extensive analyses of the device's performance. The difficult procedures that were followed are reported in the sections 3.2.3 to 3.2.6.

3.2.2 Secular equilibrium of the short lived Radon progeny

The large difference which exists between the half life of Rn-222 (3.82 days) and the half lives of its short lived daughters results in the possibility for a sealed sample of Radon to reach a situation called secular equilibrium (Appendix 1.2). The time necessary for the secular equilibrium to be reached in the case of the Radon progeny is demonstrated by an output of the program mentioned under paragraph 3.2. The initial condition consists of a pure Radon sample with a unit activity. The numbers reported in Table 3.1 and graphed in

Fig. 3.2 describe the activity of each Radon daughter as a fraction of the parent activity and as a function of time. Besides observing that the short lived daughters have practically reached the activity of Radon within 4 hours, it is of importance to note that the very long half life of Pb-210 (21.5 years) causes the remaining nuclides in the chain to have a negligible activity after 4 hours. In particular, one does not have to consider any contribution from the fourth alpha emitter, Po-210. These observations lead to the conclusion that after a minimum of 4 hours the total alpha activity of a Radon sample is, for all practical purposes, equal to three times the activity of the Radon itself. Table 3.2 shows the experimental data collected during the buildup of alpha activity. Because of the rapid growth of the second alpha emitter (Po-218) the initial activity of the sample is inferred from the one measured 15 minutes after the sample has been introduced into the scintillation chamber. This delay allows enough time to transfer the scintillation chamber from the location of sampling to the counter and also allows establishment of the reference count, which lasts 5 minutes, in a region of the buildup curve where the activity does not vary too rapidly during the counting. The extrapolation to find the initial activity of the sample is based on the theoretical evolution of the sample's activity which is predicted by an output of the DEC SER program. The maximum experimental activities expressed as a fraction of the theoretical activity at time $t = 0$ reach values which average closely to the theoretical maximum. The area of maximum activity also coincided with the one predicted by the solution of the equations for radioactive series. Discrepancies are believed to be introduced by the presence of short lived daughters collected with the sample.

The above theoretical analysis assumed that the air sample contained Radon only and no other alpha emitters. As we pointed out in explaining the discrepancies between the theoretical buildup and the values recorded in Table 3.2, this is not the case for ordinary air samples which, because of background, always contain Radon daughters. In spite of a filtration which tends to reduce drastically the amount of daughters collected in the container, Radon daughters are often introduced in the scintillation chamber at the time of sampling. The numbers presented in Table 3.3 and graphed in Fig. 3.3 demonstrate that the alpha activity of any short lived Radon daughter collected at the time of sampling is negligible at the time of sample counting, i.e., 4 hours later. The case considered in Table 3.3 is the worst possible case. It corresponds to the sampling without filter of a Radon atmosphere of unit concentration and in secular equilibrium. The total residual activity of the daughters Po-214 and Po-218 after the four hour buildup period amounts to 1.106×10^{-2} compared to 2.912 for the total activity of the Radon and the daughters formed during the same period (Tables 3.3 and 3.2).

3.2.3 Calibration. Several factors introduce a discrepancy between the net alpha activity of a sample and the actual photoactivity of the scintillation chamber. First, a fraction of the alphas will strike the glass window and result in no photo-emission. Secondly, a small fraction of the alphas striking the ZnS(Ag) will not result in a photon emission. Finally, only a fraction of the visible photons emitted by the ZnS(Ag) will be emitted in the direction of the glass window and will be recorded by the photomultiplier tube. A calibration is therefore needed to establish an overall yield Y defined as

$$Y = \frac{\text{count rate}}{\text{alpha activity of sample}} \quad (3.21)$$

In view of the analysis which was presented on the respective activities of the alpha emitters in a sample, this overall yield Y can easily be converted to a "Radon yield":

$$Y_R = \frac{\text{count rate}}{\text{pCi of Radon}} \quad (3.22)$$

The calibration of a battery of scintillation chambers requires that a few of them be calibrated first by sampling an atmosphere of known Radon content. The remaining chambers are then calibrated by simultaneously sampling an unknown atmosphere with calibrated and uncalibrated chambers aligned in series. The calibration of the reference chambers was accomplished on an experimental system developed by Dr. K. J. Schiager and his doctoral student D. R. Alexander (Department of Radiology and Radiation Biology). The procedure followed for a calibration run consisted of the following:

1. Allow a calibrated NBS (National Bureau of Standards) Ra-226 solution to build to radioactive equilibrium with its first daughter, Rn-222. The evolution of the activities of the various nuclides in the Radium solution is shown in Table 3.5. Radioactive equilibrium of Radon is seen to be achieved practically within 40 days.
2. Record a gamma spectral analysis of the solution after the equilibrium of Radon has been attained.
3. Entrain the Radon out of the solution into a large volume jug (approximately 20 liters) by bubbling air through the solution.

4. Separate the jug from the solution flask, seal both containers, and allow the residual Radon left in the solution to reach equilibrium with its daughters, while the daughters initially in equilibrium with the evacuated Radon are allowed to decay.
5. Perform a new gamma spectral analysis of the Radium solution. The ratio of the present Radon daughter activities to the ones recorded under step 2 determines the fraction of residual Radon left in the solution. Consideration has to be given to the fact that all the Bi-214 present in the solution 4 hours after the bubbling operation does not evolve only from the residual Rn-222. A fraction of the Bi-214 activity which is recorded by the second gamma spectral analysis results from series decay from the Ra-226. An output of the program in Appendix 3.1 (Table 3.4) indicates that the activity of Bi-214 in an originally pure sample of Ra-226, reaches 2.1 percent of the Ra-226 activity after 4 hours. Finally, consideration should also be given to the remaining activity of the Bi-214 which was contained in the solution before the bubbling operation. Table 3.2 indicates that in the absence of the parents Ra-226 and Rn-222, an initial unit information allows the determination of the exact activity which was removed from the solution and introduced into the jug.
6. The atmosphere of the large jug which now contains a known concentration of Radon is then sampled with the Radon scintillation chambers which are to be calibrated.

The results of one calibration run were the following:

The Ra-226 solution was calibrated by NBS in May 1968 and certified to contain 8000 pCi of Ra-226. At the time the scintillation chambers' calibration experiment was run (3/26/73), the residual activity of Ra-226 was

$$I(t) = 8000 e^{-(0.693)(4.8)/1602} = 7984 \text{ pCi Ra-226 .}$$

The last time the Radium solution had been emptied of its Radon content was on 2/16/73, i.e., 38 days earlier. Figure 3.5 indicates that after 38 days, the activity of Radon in the Radium solution has reached 99.8992 percent of the parent activity. The respective Rn-222 and Bi-214 activities in the Radium solution before the bubbling operation was therefore:

$$7984 \times 0.99892 = 7975 \text{ pCi .}$$

The comparison of the net counts found in two Bi-214 photopeaks (0.609 Mev and 1.76 Mev), indicates a reduction of Bi-214, and therefore of Rn-222, activity after the bubbling operation. Ratios of net activities in these two peaks for increasing integrated widths are reported in Tables 3.6 a and b. The different ratios show good agreement and their average indicates that the Bi-214 activity in the Ra-226 solution 4 hours after, was 4.5 percent of its activity before bubbling. This activity ($7975 \times 0.045 = 359 \text{ pCi}$), is made of three contributions: the Bi-214 generated in 4 hours by the Ra-226, the residual Bi-214, 4 hours after the removal of the Radon from the solution and the Bi-214 generated over the 4 hour equilibrium

buildup period by the Radon left in the solution after the bubbling operation. The activity of Bi-214 generated in 4 hours in the Radium solution amounts to 2.1 percent of the Radium 226 activity ($7984 \times 0.021 = 168$ pCi) while the residual fraction of the 7975 pCi of Bi-214 which were present at the time of bubbling amounts to 0.788 percent of the initially present activity ($7975 \times 0.788 \times 10^{-2} = 63$ pCi). The net amount of Bi-214 generated by the Rn-222 left in the solution after the bubbling operation is:

$$359 - 168 - 63 = 128 \text{ pCi ,}$$

and the net activity of Rn-222 removed from the Radium solution by the bubbling operation is:

$$7925 - 128 = 7847 \text{ pCi .}$$

The total volume of the system (including the circulation pump and the auxilliary tubing) had been flushed with Nitrogen before being connected to the Radium solution container and amounted to 21.54 liters. The transferred Rn-222 activity resulted in a concentration of:

$$7847/21.54 = 364 \text{ pCi/liter in the large jug .}$$

The results of the jug atmosphere sampling with Radon scintillation chambers (Nos. 4,15,31,33) are reported in Table 3.7 as well as the discrepancy recorded for each chamber between the known value of the atmosphere sampled and the values obtained by using their assumed yields. The experiment resulted in the necessity to adjust the yields of the scintillation chambers to the new values listed in Table 3.7. The other chambers used in this study were calibrated by sampling unknown Radon atmospheres with two calibrated chambers in series.

3.2.4 Data collection and analysis. After the background of the scintillation chamber (A_0 counts/m) has been recorded, the atmosphere to be evaluated with respect to Radon concentration is established in the chamber. The latter is then kept in the dark for a minimum of 4 hours. The darkness is required to avoid any activation of the chamber's coating by light. The 4 hour period allows the short lived Radon daughters to reach a state of secular equilibrium, while the Radon progeny introduced in the chamber at the time of sampling, decay to a negligible activity. The photoactivity of the chamber, $A(t)$, is then recorded and the Radon concentration at the time of sampling, $[Rn]$, is deduced from the following expressions:

$$[Rn] = \left[\frac{A(t) - A_0}{3Y_R V} \right] e^{\lambda t} \quad (\text{pCi/liter}) \quad (3.23)$$

where

λ = decay constant of Rn-222 ($7.56 \times 10^{-3} \text{ hr}^{-1}$)

Y_R = Radon yield of chamber (count rate/pCi of Radon)

V = volume of chamber (liters)

t = decay time (hours)

The factor "3" in the denominator of (3.23) takes into account that the alpha activity in the chamber equals three times the Radon activity. The exponential in (3.23) is introduced to infer the Radon activity at the time of sampling by observation of its activity at the time of counting.

Equation (3.23) allows the deduction of a Radon concentration from a count performed at any time ' t ' after sampling, provided that the elapsed time is greater than 4 hours. Nevertheless, one should avoid prolonging unnecessarily the residence time of a Radon sample in the scintillation chamber so as to reduce to a minimum the buildup of the

long lived Pb-210 and the amount of plateout by the Radon progeny on the walls of the chamber. The nuclides which plate out will not be removed by the flushing operation which the chamber experiences immediately after counting, and the plated nuclides ultimately decay to the long lived Pb-210. Pb-210, in turn, through its daughter Po-210, contributes to the alpha background activity in the chamber. Table 3.9 reproduces the activity buildup of the long lived Pb-210 and its daughter Po-210 as a function of the time spent in the scintillation chamber.

3.2.5 Short term background suppression after counting. After a gas sample has been analyzed, the scintillation chamber is flushed so as to remove the radioactive nuclides which have not plated out on the walls of the chamber. The short lived daughters attached to the walls result in a short-term background activity for the chamber. This short-term activity, which is superimposed to the long-term background of the chamber, slowly decays as the high activity, short-lived, daughters decay into the long lived, low activity, Pb-210. If the Rn-222 content of the chamber could be removed totally by the flushing operation (which could be expected for an inert element such as Radon), a reasoning similar to the one applied to the parasitic short lived daughters collected at the time of sampling, could be applied to the Po-214 and Po-218 which remain attached to the walls of the chamber after flushing. Their activity, which corresponds to the total short-term background activity of the chamber, should drop after 4 hours to a negligible quantity. As a result, a new gas sample could be collected immediately after a flushing performed in such a way that all unattached nuclides in the chamber have been evacuated. For a chamber

volume of 135 ml, a flushing flow rate of 5 liters/m for 15 minutes should suffice. While the new Radon gas sample is allowed to reach secular equilibrium (4 hours), the plated-out short lived daughters and the parasitic short lived daughters introduced at the time of sampling, would then decay to a negligible activity.

Experiments were performed to determine the validity of the above reasoning and it was found to apply, with the exception of the assumption of total Radon removal by a single high flow rate, short-time flushing.

As experiments progressed, background counts and efficiency of the instrumentation were constantly recorded. The data related to these experiments are reported in Tables 3.10 a and b. The first short-term background decay experiment is reported in Table 3.11. The steady background of the chamber used was 135 counts/5 m. An atmosphere of 500 pCi/liter was sampled and counted 10 hours later. After being counted the chamber was flushed for 30 minutes with an air pump delivering 5 liters/m and resealed to simulate the collection of a new sample. The chamber was then counted 4 hours after the flushing operation so as to evaluate the importance of the residual short-term activity which would have been present at the time of counting of the next sample. The residual short-term activity above the long term background was found to be 40 c/5 m, i.e., approximately 5 percent of the activity sampled.

A second similar experiment, involving the simultaneous monitoring of two chambers was then performed with higher concentration samples (50,000 pCi/liter and 100,000 pCi/liter). The flushing operation was continued for 10 minutes with a flow rate of 5 liters/m. The

results of the experiment are presented in Table 3.12 and show again a residual activity of 5 percent of the sample's activity after a decay period of 4 hours.

A third experiment was performed with concentrations of 30,000 pCi/liter and 75,000 pCi/liter. The flushing amounted to 5 liters/m for 30 minutes. The observation of the background decay was continued for 5 hours. As noted in Table 3.13, the background of both chambers showed a net tendency to increase after the fourth hour. One of the chambers was then flushed anew for 5 3/4 hours while the other was left sealed in the counter. After this last flushing operation, the twice flushed chamber showed a drastic drop in activity to almost steady background while the other chamber seemed to indicate a slow background increase over the last 6 hours. This last chamber was then flushed for 14 minutes (5 liters/m) which reduced immediately its activity by two thirds. Flushing of both chambers overnight finally led to activities corresponding to their respective steady backgrounds.

A fourth experiment was performed with concentrations of 40,000 pCi/liter and 65,000 pCi/liter to observe the decay of background of a chamber that was continuously flushed. While one chamber was flushed during the whole experiment, the second was flushed during only the first hour and then sealed. In the region where the count is sufficient to provide reliable figures, the results tabulated in Fig. 3.14 show a good agreement for the continuously flushed chamber between the predicted theoretical decay and the counts registered. The results for the chamber which was flushed for only one hour confirmed the tendency for the background to increase after 4 hours.

It is concluded from the data collected in the above experiments that the observed behavior of the scintillation chambers matches the predicted values during the short-term background decay period provided that the chamber is flushed during the entire decay period. The drastic reduction in activity obtained by a second flushing performed several hours after the first one (third experiment), indicates that the presence of airborne nuclides in the chamber exist four hours after the first flushing. This presence strongly suggests that the parent Radon diffuses through the ZnS(Ag) coating of the chamber while the high concentration sample is in the chamber, but diffuses back into the chamber volume after the first flushing operation reduces the Radon concentration in the chamber to a negligible value. The slight tendency of the activity of the sealed chamber to increase as the Radon diffuses out of the coating could stem from the fact that a proportionally larger activity of Radon is entrapped in the walls than is plated on the walls under the form of individual short lived daughters. After removal of the airborne Radon and progeny, a new equilibrium has to be achieved between the Radon left in the walls and the progeny attached to the walls. The buildup to a new equilibrium results in a slow increase of the chamber's activity.

3.2.6 Reproducibility of the measurements. In order to establish the reliability of the measurements obtained with Radon scintillation chambers, seven measurements of unknown Radon concentrations were made with the same four Radon scintillation chambers placed in series. These measurements were made before the final yields of the respective chambers were established. The acceptability of the measurements was evaluated by the consistency with which the data collected in

each measurement would scatter around the mean value obtained for all four chambers. The deviations from the mean are reported in Table 3.8 for each chamber.

3.3 Conclusions

The use of the Radon scintillation chambers that were built by the project experimenters as a means of recording Radon concentrations was shown to be a reliable technique since the behavior of the instrumentation reproduces the data predicted by the solutions of the equations describing the time evolution of naturally occurring radioactive chains. Backgrounds and efficiencies of the counters must be constantly verified before recording data to compensate for the drift of the instrumentation. Special care must be devoted to the protection of the instrumentation and of the chambers from the light. The residence time of radioactive samples in the chambers must be kept to a minimum (4 to 5 hours) so as to reduce the buildup of the steady background to a minimum and the flushing operations should be performed over long periods of time (4 hours) rather than with a high volumetric flow rate applied over a short period of time. If, in order to collect a new sample, a short thorough flushing is applied, the residual short-term activity above the steady background of the chamber will amount to approximately 5 percent of the activity of the preceeding sample at the time of counting of the next sample. Finally, the manipulations and calculations required for the calibration of scintillation chambers by means of Radon atmosphere extracted from a standard Ra-226 solution by a bubbling operation have been shown to give satisfactory results.

4. PHYSICAL CHARACTERISTICS OF URANIUM MILL TAILINGS AND CONCRETE

The quantization of analytical models aimed at predicting the gamma-ray and Radon progeny exposures in structures contaminated by Uranium mill tailings requires the determination of the physical parameters relevant to Radon diffusion and which characterize the media involved, i.e., the tailings and the concrete.

The characterization of the Uranium mill tailings requires not only an assay of the Radium content of the tailings but also an evaluation of the amount of Radon released by these tailings. Experimental evidence shows that only a small fraction of the Radon born from the decay of Radium is free to diffuse. Following the terminology adopted by the literature [6.10], this fraction will be called the "emanation power" of the tailings. The remainder of the Radon is entrapped in the lattice of the sand particles which constitute the tailings. Therefore the measurable Radium activity of the tailings needs to be corrected by a factor representing the fraction of free Radon in order to assess the true magnitude of the Radon source constituted by a certain amount of tailings.

The physical characteristics of concrete were also analyzed with emphasis on the features relevant to the diffusion of Radon.

4.1 Radium Assay of Uranium Mill Tailings

It was the purpose of this experiment to assay the Radium content of recent tailings (sand fraction) from the Climax Uranium mill in Grand Junction and old tailings from the Union Carbide mill in Uravan. The Radium contents of the tailings samples were compared to the

Radium content of ordinary topsoil sampled on the campus of Colorado State University.

4.1.1 Experimental procedure. Samples of the source material to be assayed were sealed in plastic Petri dishes (150 mm diam., 25 mm deep) with a clear air drying varnish (1202 Glyptal). Seven samples of the Grand Junction tailings and one sample of ordinary topsoil weighing 500 g net each and two samples of the Uravan tailings weighing 400 g each were prepared. The lower density of the Uravan tailings did not allow the Petri dishes to contain more than 400 g.

In order to achieve similar geometries and counting efficiencies, a standard Radium source was prepared which reproduced the configuration of the samples. A solution of 8000 pCi of Ra-226 was obtained from the National Bureau of Standards. The solution amounted to 20 ml and contained some BaCl_2 (0.2% by weight) and HCl (4.5% by weight). This solution and 30 ml of HCl used as a rinse to remove all of the radioactive solution from its shipping vial were neutralized by 7.5 ml of $\text{NH}_4(\text{OH})$. This neutral solution was then incorporated into 230 g of Elmer's glue and sealed in a Petri dish identical to the ones containing the material to be assayed. The glue was used to distribute the standard Radium solution over a thickness comparable to that occupied by the tailings samples. The neutralization of the acid was required in order to avoid a reaction which drastically darkened the Elmer's glue. A blank sample containing only Elmer's glue was prepared in order to evaluate the background counting rate of the instrumentation.

4.1.2 Results analysis.

a. Bi-214 photopeak at 1.76 MeV. Ratios of the integrated 1.76 MeV photopeaks observed from the tailings samples and the standard

source provided a measure of the actual Radium content of the samples. Background and standard source readings, normalized to 10 n counts, are reported in Table 4.1 along with the resulting net count rate of the standard source. The readings obtained with the various samples are presented in Table 4.2 where the first seven lines correspond to Grand Junction tailings, the next to lines to Uravan tailings and the last line to ordinary topsoil. The measured Radium concentrations in the various samples with identical origin show good agreement and the average values amount to 0.12 g Ra-226/1000 dry metric tons for the Grand Junction tailings, 0.21 g Ra-226/1000 dry metric tons for the Uravan tailings and 10^{-4} g Ra-226/1000 dry metric tons for ordinary topsoil.

b. Spectrum integrals from 0.5 to 2 MeV. If one assumes that radionuclides outside the Ra-226 chain, e.g., K-40, contribute very little to the spectrum of the tailings, a comparison of the integrated spectra should also provide a reliable estimate of the samples' Radium content. Tables 4.3 and 4.4 show the analysis which was performed on the K-40 characteristic 1.46 MeV photopeak. The ratios of this K-40 photopeak activity in the tailings samples to its activity in the standard source are very similar to the ratios found in the case of the 1.76 MeV photopeak of Bi-214 (Table 4.2). The resulting averages were 7.38 vs. 7.49 for the Grand Junction samples and 13.34 vs. 13.37 for the Uravan samples. It was concluded that the relative importance of the K-40 contributions to the tailings spectra was negligible. A comparison of the integrated spectra from the standard source and from the tailings samples should therefore provide a valuable estimate of the tailings' Radium content based on a broader source of data than the single 1.76 MeV photopeak. One can observe this is is not

the case for the sample of ordinary topsoil since its K-40 (1.46 MeV) photopeak is 1.15 times more active than the standard source's same spectrum region, compared to a ratio of only 0.075 in the case of the Bi-214 (1.76 MeV) photopeak.

The comparison of the spectra integrated over the range 0.5 to 2 MeV is presented in Tables 4.5 and 4.6 following a pattern identical to the one adopted for the analysis of the 1.76 MeV photopeak. The average Radium contents of the tailings samples amount to 0.13 g Ra-226/1000 dry metric tons for the Grand Junction tailings and 0.23 g Ra-226/1000 dry metric tons for the Uravan tailings. These results agree with the 1.76 MeV photopeak analysis. In the case of ordinary topsoil, the relative activity of the integrated spectrum is higher than the one from the 1.76 MeV photopeak. This result was expected on the basis of the analysis which revealed a greater relative importance of the K-40 contribution in the ordinary soil as opposed to a negligible relative contribution in the tailings samples. Table 4.7 summarizes the results of the different assays.

4.2 Emanation Power of Uranium Mill Tailings

4.2.1. Experimental procedure. Four cylindrical metal containers (two large: 11 3/4 in. diam. and 13 1/4 in. height and two small: 5 3/8 in. diam. and 5 5/8 in. height) were sealed after being completely filled with well compacted dry tailings. One container of each size (No. 1) was filled with pure tailings while the other was filled with tailings which had already been used for construction purposes and contained rocks and pieces of concrete. The lateral surface of each container was equipped with two diametrically opposed air sampling outlets located at half height. The tailings were left in the sealed

containers during thirty days for the Radon to reach an equilibrium with the Ra-226 (Table 4.8). Samples of the gas phase of the tailings were then collected with Radon scintillation chambers and their Radon concentration evaluated.

4.2.2 Theoretical foundations. The measure of the emanation power (E) of the tailings by the present method, relies on a balance equation of the Radon population in the gas phase of the tailings. After radioactive equilibrium has been reached, the Radon concentration in the container, and in particular in the gas phase of it, remains constant at a level such that:

$$[\text{Production rate}]_{\text{gas phase}} = [\text{Decay rate}]_{\text{gas phase}} \quad (4.1)$$

Evaluation of each term in (4.1) leads to the following expressions (in atoms of Radon/cm³-s):

$$[\text{Production rate}]_{\text{gas phase}} = V_t \times \rho \times [\text{Ra}] \times 3.7 \times 10^{10} \times E, \quad (4.2)$$

where V_t = total volume of tailings (cm³)

ρ = density of tailings (g/cm³)

[Ra] = Radium content of tailings (g Ra-226/g tailings)

E = emanation power of tailings.

Also,

$$[\text{Decay rate}]_{\text{gas phase}} = V_t \times \text{port} \times C_o \times \lambda, \quad (4.3)$$

where port = porosity of the compacted tailings

C_o = sampled Radon concentration (at/cm³)

λ = Radon decay constant (s⁻¹) .

Substitution of (4.2) and (4.3) into (4.1) provides the following expression for E:

$$E = \frac{\text{port} \times \lambda}{3.7 \times 10^{10} \times \rho \times [\text{Ra}]} \times C_o \quad (4.4)$$

If C_o is expressed in pCi/liter, a transformation of (4.4) is necessary through the following conversion:

$$C'_o \text{ (pCi/liter)} = \frac{C_o \text{ (at/cm}^3\text{)} \times \lambda \times 10^5}{3.7} \quad (4.5)$$

This transforms (4) into

$$E = \frac{\text{port}}{\rho \times [\text{Ra}] \times 10^{15}} \times C'_o \quad (4.6)$$

where C'_o = sampled Radon concentration (pCi/liter).

The value of $[\text{Ra}]$ was established in section 4.1.2. The present method of evaluation of E requires that ρ and port also be established to evaluate (4.6).

4.2.3 Density and porosity of Uranium mill tailings. Aside from the importance of the parameter ρ in computing (4.6), other instances require that its value be known. The Radium content of the tailings has been established in a previous experiment and was stated in terms of grams of Radium per thousand dry tons of tailings. The amounts of tailings used in our experimental facilities (experimental modules and Radon barrier testing facilities) are known in terms of volumes occupied. A proper evaluation of the Radium present in various locations requires, therefore, an estimate of the tailings density.

The density of the tailings was determined in cases of minimum and maximum compaction by weighing 200 cm³ and 500 cm³ samples.

The least compaction was achieved by slowly filling a graduated container with tailings while avoiding either dropping the tailings from any appreciable height or shaking the container. The experiment was repeated seven times with a 200 cm^3 volume and three times with a 500 cm^3 volume in order to assess if the tailings' own weight would influence the measurement. The 500 cm^3 data showed a slight but negligible increase over the 200 cm^3 data. The data are reported in Table 4.9.

The maximum compaction was obtained by two different methods. The first one consisted of tapping the walls of the container and pounding it against a hard surface. This operation was repeated with three samples, each one occupying a final volume of 500 cm^3 . The second method for achieving maximum compaction consisted of saturating a known height of dry, loose tailings with water. This proved to considerably reduce the volume occupied by the loose tailings (from 200 cm^3 to 180 cm^3). The densities of compacted tailings obtained with both methods are reported in Table 4.10.

Because of the method followed, an estimate of the porosity was only obtainable in the case of compacted tailings. This is probably the closest situation to the one encountered in practical applications. During the compacted tailings density measurements, a record was kept of the amount of water required to saturate a known final volume (180 cm^3) of compacted tailings. The ratio of the water volume used to the final volume of the tailings was assumed to represent the compacted tailings porosity. The experimental results are reported in Table 4.11.

In conclusion, the average density of noncompacted sand was found to be 1.45 g/cm^3 and the compaction operations, either mechanical or hydraulic, resulted in an average compacted tailings density of 1.62 g/cm^3 . This amounts to an average increase of 11.7 percent in density after compaction.

The average compacted tailings porosity was found to be 36.6 percent. This figure is in good agreement with the value of 40 percent which is accepted for all practical purposes in the case of sand.

4.2.4 Emanation power. Results and discussion. The parameters port , ρ , and $[\text{Ra}]$, which were established in previous experiments, amount to the following values:

$$\text{port} = 0.36 \quad (4.7)$$

$$\rho = 1.62 \text{ g/cm}^3 \quad (4.8)$$

$$\begin{aligned} [\text{Ra}] &= 0.125 \text{ g Ra-226/1000 tons tailings} \\ &= 0.125 \times 10^{-9} \text{ g Ra-226/g tailings} \end{aligned} \quad (4.9)$$

Insertion of (4.7), (4.8) and (4.9) into (4.6) provides

$$E = 1.8 \times 10^{-6} C'_0 \quad (4.10)$$

with C'_0 in pCi/liter.

Using the recorded values of C'_0 presented in Table 4.12 for the pure tailings, one determines the following average value of E :

$$E = 0.18 \times 10^{-5} \times 127,000 = .2286 \approx 20\% \quad (4.11)$$

The above analysis has led to an expression of E (4.6), which is logically independent of the volume of the container (V_t). Yet, the experimental method described here does not allow an accurate measure of C'_0 unless the volume and the distance between the sampling outlets of the container are greater than a certain minimum. For

values larger than these practical minima, experimental evidence does support the independence of the measured value of E from the container's size. For containers which are too small, the observed value of C'_0 is not the one existing in the gas phase of the tailings because of the dilution taking place when the tailings' atmosphere is mixed with the total volume (V_s) of the sampling apparatus (scintillation chamber, pump and associated tubing). In order to reduce this dilution to a minimum, the product (port $\times V_t$) should at least be one order of magnitude larger than V_s . Furthermore, an observation of the flow pattern of the circulation induced by the sampling operation reveals that only a fraction of the total available volume (port $\times V_t$) is entrained by the pump (Fig. 4.1). The inaccessibility of an unknown fraction of the container's volume precludes any accurate correction for the dilution effect and requires that the geometry of the operation and the location of the sampling ports be such that the "affected volume" be at least one order of magnitude larger than V_s . Table 4.13 reproduces the data obtained with the smaller containers. The values of C'_0 are, in this case, equal to one half of the values recorded with the larger containers. This results from the fact that the volume of the sampling device was 145 ml and the gaseous volumes of the containers were respectively, 752 ml and 9192 ml. Furthermore, the affected volume increases with the distance between the sampling ports. The assurance that the larger containers were large enough was gained from other experiments on Radon sealants, when even greater quantities of tailings were sealed. The concentrations sampled out of these large volumes did compare with the ones presented in Table 4.12.

4.3 Physical Structure of Concrete

The observed importance of the concrete layer in reducing the Radon flux and in calculating the gamma exposure increase as a result of sealant applications led us to perform a study of the nature, structure and origin of the concrete porosity. The study is presented in Appendix 4.1 and can be summarized as follows with respect to the problem of Radon diffusion in concrete: The cement paste in concrete [4.1, 4.5] occupies from 23 percent (lean mix) to 36 percent (rich mix) of the total volume, the sand 25 percent to 30 percent and the aggregates the remainder. The overall porosity of concrete will, consequently, depend as much on the porosity of the cement paste as on the aggregates' porosity. It will always be less than the volume-weighted average of these two components due to the presence of sand in the mix.

The porosity of the cement paste consists of two types:

1. The gel porosity results from the gel-like structure adopted by the hydration products of the dry cement. The pores of this gel structure constitute 28 percent of the hydrated paste and are filled with entrapped water molecules. The pores range in size from 1.5 to 8.0×10^{-7} cm [4.5, 4.3]. Their ability to allow Radon diffusion across the concrete would be very small and evidence has been presented of their sieving effect on gas molecules such as Nitrogen, Hydrogen and Helium [4.4].
2. The capillary porosity results from the net reduction in volume of the water-dry cement mixture during the hydration process. The capillary pores are much larger in size than the gel pores and although their diameter distribution centers

around 2×10^{-5} cm [4.6] it can extend up to 1.3×10^{-3} cm [4.3].

Immediately after mixing of the cement paste, the capillary porosity is considered to be made of, and filled by, all the water which surrounds the dry cement grains. As the hydration of the cement grains proceeds, the capillary system is progressively replaced by gel pores. The water to cement ratio by weight (w/c) used in the mix dictates the amount of capillary porosity left in the paste after all the cement has been hydrated. When a paste involving an optimum w/c = 0.42 is cured under dry conditions (i.e., without additional supply of water) there always results a minimum capillary porosity of about 8 percent. If the w/c ratio is increased, which is often the case on site, the capillary porosity increases also. As the curing of the cement paste proceeds, the capillary pore system becomes segmented by the gel structures unless the w/c ratio exceeds 0.7, in which case the segmentation will never occur. According to the calculational methods presented in Appendix 4.1, a w/c ratio of 0.7 results in a capillary porosity of 33.3 percent.

The porosity of the common aggregates ranges from a fraction of a percent to 20 percent for dense stones and can reach 50 percent for lightweight aggregates [4.3]. Frequently, the pores in the aggregate are at least the size of the paste capillaries. The role of aggregates in allowing Radon to diffuse through concrete is important.

It is concluded that the type of aggregates used in the concrete and the w/c ratio used in the mix are the ruling parameters in deciding if a concrete could be very permeable to Radon. The overall concrete porosity which should be considered as relevant to the

problem of Radon diffusion can consequently range from a few percent (optimum mix and dense aggregate) to about 25 percent (high w/c ratio and porous aggregates).

5. THE EFFECTIVE DIFFUSION COEFFICIENT OF RADON IN POROUS MEDIA

5.1 Need for a Quantitative Description of Radon Diffusion

The application of a sealant on the indoor face of a concrete wall adjacent to a layer of tailings emanating Radon gas, results theoretically in an increase of the equilibrium Radon concentration in the tailings and throughout the treated concrete wall. In a steady state situation, this increase is accompanied by a proportional increase in the Radon progeny concentration which, in turn, leads to an increase in the gamma radiation emitted from the wall and from the tailings. An evaluation of the order of magnitude of this increase, requires that the Radon concentration profiles in the tailings and concrete walls be established before and after the application of the sealant. The profiles are then used in shielding calculations involving a multilayered source with variable source strength. The various profiles necessary for the shielding calculations result from the solution of a system of equations describing the diffusion processes taking place throughout the three media involved: soil, tailings, and concrete. The present chapter deals with the derivation of the applicable equations of diffusion in these media and with the evaluation of the parameters necessary to generate quantitative information. The validity of the model and the accuracy of the measured parameters will then be analyzed by comparing their combined prediction to an actual situation.

5.2 Effective Diffusion Coefficients and Linear Diffusion Model

5.2.1 Types of diffusion. The concentration profiles resulting from a diffusion process, are derived from a balance equation applied to a control volume of the considered medium. The most general form of the balance equation used in this work is derived for the tailings, which, besides being porous, also contain the Radon source term. The cases treated in the present study involve one dimension only, and if we consider a control volume of porous material limited by two unit surface areas perpendicular to the axis and separated by a distance 'dx', we have:

$$F|_x - F|_{x+dx} - Ddx + S'dx = 0 \quad (5.1)$$

where $F|_x$ = flux into the control volume at x (at/cm²)

D = decay rate of the diffusing gas in the C V (at/cm³)

S' = production rate of the diffusing gas in the C V
(at/cm³)

On the basis of experimental evidence presented further in this chapter, the problem can be considered as a diffusional flow under constant total pressure. The net flow of the gas results only from the thermal agitation coupled to a concentration gradient. In the case of gases, this is equivalent to a partial pressure gradient. With the total pressure remaining constant throughout the system, there must exist an equivalent counterflow of air, leading to a zero net flow rate. In such a case, the evaluation of the flux terms in the diffusion balance equation results from Fick's law for diffusion [5.1]:

$$F(x) = -k_o \frac{dC(x)}{dx} \quad (5.2)$$

where k_0 = diffusion coefficient (cm^2/sec)

$C(x)$ = concentration in the gas phase (at/cm^3).

Since F is defined as the flow rate per unit cross section area, the diffusion coefficient k_0 represents the flow rate ($\text{at}/\text{cm}^2 \text{ sec}$) which takes place across a unit section of the control volume in the presence of a unit concentration gradient. Fick's law was established experimentally for diffusion of substances in homogeneous media. Its application to porous media, where the diffusion takes place in the voids formed by a solid matrix, requires a careful definition of the concepts used. The lack of such precaution, in the literature that was surveyed, has revealed a use of identical symbols for different physical quantities. The consequences of such practices becomes apparent when numerical values are needed for the parameters.

In the case of concentration driven diffusion processes, Fick's law, with a diffusion coefficient defined as the flux through a unit section of the material in the presence of a unit concentration gradient is commonly applied to materials covering the complete range of compaction that could possibly be encountered. The classification of these various degrees of compaction and their associated effective diffusion coefficients can be described by the following succession:

Consider a cylinder of unit cross sectional area and the diffusion which takes place along its axis. If the cylinder is filled with air and contains a higher Radon concentration at one end, diffusion will take place according to

$$F(x) = -k_0 \frac{dC(x)}{dx}$$

where F = mass flow rate across section at x

k_0 = diffusion coefficient of Radon in air

$C(x)$ = Radon concentration at x .

If, in presence of the same concentration gradient, the cylinder is filled with a solid matrix of porous material, the diffusion of Radon will take place at a slower rate as the Radon atoms progress through the tortuous maze of capillaries. The flow can, nevertheless, be described by an expression of the form (5.3), where k_0 is replaced by an effective diffusion coefficient k_e , smaller than k_0 , and where the concentration $C(x)$ is measured in the gas phase of the porous medium. This results in Fick's law being written as

$$F(x) = -k_e \frac{dC(x)}{dx} \quad (5.4)$$

Expressions have been developed to predict the magnitude of k_e on the basis of the internal structure of the porous material and the relative size of the pores with respect to the mean free path of the diffusing substance in the capillary filling medium [5.1, 5.2, 5.4]. When the capillary diameter is much larger than the mean free path 'l' of the diffusing substance, the diffusion is said to be purely molecular since relatively little interaction occurs between the walls of the capillaries and the diffusing substance. A general form of k_e is then [5.2]:

$$k_m = \frac{p}{q^2} k_0 \quad (5.5)$$

where p = porosity of the medium

q^2 = tortuosity factor = L_e/L

L = linear dimension of the medium

L_e = effective length travelled by molecules to cover a distance L .

When the pore diameters are much smaller than '1', the diffusion process in which the diffusing molecules collide mainly with the walls is called "Knudsen diffusion." As opposed to the molecular diffusion case, the diffusion of a substance through the capillaries of the porous medium becomes independent of the substance filling these pores. An example of expression for k_e is then:

$$k_K = 9.7 \times 10^{-3} \bar{r} \sqrt{\frac{T}{M}} \quad (5.6)$$

where $\bar{r}$ = radius of the pores (cm)

T^0 = temperature in degrees Kelvin

M = molecular weight of diffusing substance (g mass/g mole).

When '1' and the diameter of the pores are of the same order of magnitude then transition or mixed type diffusion occurs. In such cases, an expression of k_{tr} which was derived on the basis of a momentum balance [5.2], reads:

$$k_{tr} = \frac{1}{\frac{1-\alpha X_A}{k_m} + \frac{1}{k_K}} \quad (5.7)$$

where $\alpha = 1 + N_A/N_B$

N_B, N_A = molar fluxes of the two species considered

X_A = mole fraction of substance A.

For a system under constant total pressure, an assumption of equimolecular counter-diffusion can be made. This leads to $\alpha = 0$ and (5.7) becomes

$$k_{tr} = \frac{k_m \cdot k_K}{k_m + k_K} \quad (5.8)$$

Finally, if the cylinder is occupied by a very dense material, such as a metal, the general form of the molecular diffusion model is again applicable and use is made of an effective diffusion coefficient representative of the substance's ability to pass through the material.

The presentation of Eqs. (5.4) to (5.8) was not made with the intent of deriving values of k_e for the media studied here. They were rather introduced to emphasize that even though all the diffusion processes considered here can be considered to depend linearly on the concentration gradient, the differences in diffusion mechanisms which are expected to occur in media as different as compacted tailings and concrete, will lead, as will be shown, to very different values of k_e . The present section also provides an unequivocal definition of the effective diffusion coefficient k_e in order to insure a correct derivation of the balance equation.

5.2.2 The balance equation and the relaxation distance. According to the values determined in Chapter 4 (Physical Characteristics of Uranium Mill Tailings), the tailings used in our studies are characterized by a density $\rho = 1.62 \text{ g/cm}^3$; a porosity, when compacted on site, of $\text{por} = 0.36$; a Radium content of $[\text{Ra}] = 0.130 \times 10^{-9} \text{ g Ra-226/g}$ of tailings, and an emanation power $E = 0.20$. The control volume introduced with equation (5.1) holds $([\text{Ra}] \cdot \rho \cdot dx) \text{ gr}$ of Ra-226 and liberates $([\text{Ra}] \cdot \rho \cdot dx \cdot 3.7 \times 10^{10} \cdot E)$ atoms of Rn-222 per second in the gas phase of the tailings control volume. This corresponds to a source term:

$$S' = [\text{Ra}] \cdot \rho \cdot \lambda \cdot 10^{12} \cdot E \cdot dx \text{ (pCi Rn/s)} \quad (5.9)$$

where λ = decay constant of Rn-222 (s^{-1}).

The decay rate of Radon in the control volume dx is proportional to the total population of Radon in the gas phase of the tailings. The concentration described being the one in the gas phase, and the gas phase's volume contained in the control volume being $\text{port} \cdot dx$, we have:

$$D = \lambda \cdot C(x) \cdot \text{port} \cdot dx \text{ (pCi Rn/s)} \quad (5.10)$$

The flux across the sections at x and $x+dx$ are represented by Fick's law:

$$F(x) = -k_t \frac{dC(x)}{dx} \quad (5.11)$$

where k_t = effective diffusion coefficient of Radon in tailings (cm^2/sec)

$C(x)$ = Radon concentration in the gas phase of the tailings (pCi/cm^3).

Substitution of expressions (5.9), (5.10) and (5.11) in the balance Eq.(5.1) leads to an equation, the limit of which, for dx tending to zero, is

$$\frac{k_t}{p} \frac{d^2 C(x)}{dx^2} - \lambda C(x) + \frac{[Ra] \cdot \rho \cdot E \cdot \lambda \cdot 10^{12}}{\text{port}} = 0 \quad (5.12)$$

where $C(x)$ is expressed in pCi/liter .

In the case where Radon gas emanates from Radium bearing tailings and diffuses through soil or concrete, the source term is considered negligible by comparison with the concentrations involved and the diffusion process is then described by

$$\frac{d^2 C(x)}{dx^2} - \frac{\lambda C(x)}{k_e/p} = 0 \quad (5.13)$$

It is of importance to emphasize that Eqs. (5.12) and (5.13) should be considered as the only valid ones to represent the diffusion of a

radioactive gas through a porous medium in the most generally accepted case where:

- 1) the effective diffusion coefficient is defined as the flux per unit cross section of the porous medium (bulk diffusion coefficient).
- 2) the medium is viewed as a solid matrix containing a maze of capillaries considered to be solely responsible for the diffusion taking place (since the diffusion through the solid matrix is found to be negligible or even nonexistent).

Equations similar, but not identical to Eq. (5.13), have been used to describe the diffusion of Radon in soils [5.10, 5.15]. The difference between the version presented here and the ones just mentioned involves the omission of the porosity p . The consequences of such omission will be discussed in the next section.

The solution of (5.13) in the case of a diffusion from a plane source (C_0) in a porous medium, without sources, is:

$$C(x) = C_p e^{-\sqrt{\frac{\lambda p}{k_e}} \cdot x} \quad (5.14)$$

The inverse of the coefficient of x in the exponent is the distance necessary for the radioactive gas concentration to drop by a factor e (2.7182) and will be designated in this work as the "relaxation distance":

$$R = \sqrt{\frac{k_e}{\lambda p}} \quad (5.15)$$

where p = porosity of the medium.

k_e = effective diffusion coefficient of Radon in the medium (cm^2/s)

As will be shown later, the concept of relaxation distance is very helpful in comparing values of "diffusion coefficients" quoted in the literature, but which do not correspond to the same physical concept.

5.2.3 Effective diffusion coefficients for porous media: soil

(k_s) and tailings (k_t) as quoted in the literature. In a review article about Radon migration in the ground [5.10], Tanner presents the solution to the diffusion from a plane source (C_p) in an infinite porous medium without sources by the equation

$$C(x) = C_p e^{-\sqrt{\frac{\lambda}{D}} \cdot x} \quad (5.16)$$

where D is called a "diffusion coefficient" of Radon through soil.

Equation (5.16) results from a balance equation written as

$$\frac{d^2 C(x)}{dx^2} - \frac{\lambda}{D} C(x) = 0 \quad (5.17)$$

Comparison of Eqs. (5.16) and (5.17) with (5.14) and (5.13), respectively, leads to the interpretation of D as k_e/p where k_e is the parameter we defined as the diffusion coefficient of the porous medium.

In making use of the data found in Tanner's reviews, one should realize that the interpretation of the "diffusion coefficient" found in (5.16) and (5.17) leads to higher values than the ones corresponding to k_e . Nevertheless, the relaxation distances derived from values of D by

$$R = \sqrt{D/\lambda} \quad (5.18)$$

should compare with the ones obtained with values of k_e by Eq. (5.15). The interpretation of the diffusion coefficient found in Eqs. (5.16) and (5.17) leads to higher values of the parameter D than the ones corresponding to k_e because it accounts for the fact that the openings of the capillary system at the surface of the control volume (CV) do not cover the total area of the control surface (CS). The reference flux under a unit gradient of concentration across the control surface is then evaluated per unit surface area of pore

openings, which leads to a higher value than k_e . Provided that the porous medium is structured in such a way that any section across it reveals a ratio of open pores surface to the total surface equal to the porosity p , this last approach to the definition of the diffusion coefficient leads to a balance equation (17) which is free of the porosity parameter in the case of a medium without sources. Yet, if sources are present and feed the diffusing gas into the capillary system, the balance equation still has to consider the porosity p of the medium. This point will be developed because of the reference which will be made to a study by Kraner [5,9] which presents a balance equation with a source term and also uses the symbol D for the diffusion coefficient while defining it the way we defined k_e .

If the concept of flux per unit area of open pores is used in defining the diffusion coefficient, the balance equation can be best established for a unit volume of capillary space rather than for a unit volume of porous medium. In a one dimensional model, the control surface perpendicular to the direction of flow covers an area $1/p$ so that it contains a unit surface area of open pores. The control volume still has a thickness dx and therefore a volume $\frac{dx}{p}$ with a gaseous volume dx . The various terms in the balance Eq. (5.1) become:

$$S' = [Ra] \cdot p \cdot \lambda \cdot \frac{dx}{p} \cdot E \cdot 10^{12} \quad (5.19)$$

$$D = \lambda C dx \quad (5.20)$$

$$F(x) = -D \frac{dC(x)}{dx} \quad (5.21)$$

and the ensuing balance equation is

$$D \frac{d^2 C(x)}{dx^2} - \lambda C(x) + \frac{Ra \cdot \rho \cdot E \cdot \lambda \cdot 10^2}{p} = 0 \quad (5.22)$$

Substitution of the value $D = k_e/p$ into (5.22) transforms it into our original Eq. (5.12). The use of a diffusion coefficient D defined on the basis of the flux per unit surface of open pores, in the case of diffusion in a medium with sources, forces one to assume the equivalence of the ratio of open pore space to total control surface and the porosity p if one wishes to avoid introducing a second parameter in addition to p . The same assumption has to be made in matching the fluxes between porous media of different porosities as the solution to our problem will require. Thus, a definition of the diffusion coefficient k_e , based on the flux through the total material surface of the medium, is more reasonable and was adopted in this work. Quantitative data on the ability of Radon to diffuse through various media was obtained by computing the relaxation lengths through the appropriate formulation, i.e., (5.15) or (5.18). The values of D reported by Tanner are presented in Table S.1. The corresponding relaxation distances were computed using (5.18) and are reported in the same table.

In the same reference [5.9], but in another article studying the effusion of Radon from the ground, Kraner et al., presented the equation for the diffusion of Radon through the ground as

$$\frac{D}{S} \frac{d^2 C(x)}{dx^2} - \lambda C(x) + \beta = 0 \quad (5.23)$$

with Fick's law written as

$$J = -D \frac{dC}{dx}$$

Along with Eqs. (5.23) and (5.24), the following definitions were provided:

J = Radon flux across a spatial area or total section of the medium ($\text{Ci}/\text{cm}^2 \text{ sec}$)

D = bulk diffusion coefficient for Radon through the volume of the porous medium (cm^2/s)

C = concentration in the interstitial gas (Ci/l)

x = linear dimension of the bulk medium under consideration

β = a constant related to the emanation power of the medium into the interstitial volume of the soil.

S = porosity of medium

The definition adopted for D in Eq. (5.23) makes it obviously different from the parameter D used by Tanner, but identifies it to our k_e and, under the condition that the porosity was indeed included in the last term of Eq. (5.23), as it should, there is agreement between Kraner's Eq. (5.23) and our Eq. (5.12).

The value obtained by Kraner for D/S or k_e/p by matching a linear diffusion model to the Radon concentration profile in the ground, is $3.6 \times 10^{-2} \text{ cm}^2/\text{sec}$. This value corresponds to a relaxation distance of 131 cm and agrees with the values reported by Tanner (Table 5.1).

In his monograph on Air Chemistry and Radioactivity [5.15], Junge presents the equation describing the diffusion of radioactive gases through soils as

$$d \frac{d^2 C_s(x)}{dx^2} - \lambda C_s(x) + a = 0$$

where d = diffusion constant (cm^2/s)

a = rate of production within the soil ($\text{at}/\text{cm}^3 \text{ s}$)

C_s = concentration of diffusing gas in the soil air (at/cm^3).

Again, no mention is made of the contents of the term "a" in terms of porosity or emanation power and the absence of the porosity from the equation is explained by the assumption made that "if the soil is

sufficiently porous, diffusion proceeds as if the soil were absent."

In such a case it would be expected that the value used for d in the calculations would be the accepted value for the diffusion of Radon in air, i.e., $10^{-1} \text{ cm}^2/\text{s}$ [5.10, 5.16, 5.17]. Yet the value used for d is $5 \times 10^{-2} \text{ cm}^2/\text{s}$. This implies that the diffusion was not considered to be completely free, and that d had to be reduced to half its value in order to represent the effect of the soil in slowing down the diffusion process. The relaxation distance in this case amounts to

$\sqrt{\frac{5 \times 10^{-2}}{2.1 \times 10^{-6}}} = 154 \text{ cm}$, and still rates high by comparison with the values listed by Tanner (Table 5.1).

Several values of the effective diffusion coefficient of Radon in sand are also reported by Tanner (Table 5.1). Among these values, the first one (5.4×10^{-2}) applies to a sand with properties very similar to our radioactive tailings: $\rho = 1.4 \text{ g/cm}^3$ and $p = 0.39$. When the gas phase of the sand is more humid, the value of k_e drops as low as 5×10^{-3} (17 percent humidity).

5.2.4 Effective diffusion coefficient of Radon through concrete.

The values of Radon diffusion coefficients in soils and sands are found in the literature because of the interest in the environmental radiation background and the prospect for locating Uranium deposits based on Radon emanation rates. We were not able to find similar values for the diffusion of Radon through concrete. A direct measure of the diffusion coefficient k_c of Radon through concrete was performed with the ordinary concrete used to build our experimental facilities. The experimental measure of k_c is presented in the following section.

5.3 Experimental Measure of k_c

5.3.1. Experimental setup. The effective diffusion coefficient of Radon in concrete was measured on the basement wall of an experimental building built to perform studies on Radon progeny control. The structure (10' x 20'), was built with a basement depicted in Fig. 5.1. Uranium mine tailings were used as a fill under the basement floor slab and as a backfill around the foundations. The concrete mix formulation was provided by the contractor (Kiefer Concrete, Fort Collins, Colorado) and amounts to the following composition per cubic yard: 1800 lbs of 3/4" rocks, 1360 lbs of sand, 470 lbs of cement and approximately 26 gallons of water. The arrangements used to record Radon fluxes are shown in Fig. 5.2. In order to monitor possible pressure gradients occurring during the flux measurements, a 3/4" hole was drilled through the wall 40" above the basement floor and a 3' long metal pipe was secured in the opening. The metal pipe was inserted 6" into the tailings and was surrounded with silicone caulking over the total length of the penetration in the wall. A water manometer was then connected between the pipe and the basement atmosphere. To measure the Radon flux seeping out of the wall, a square metal container (4' x 4' x 1") was sealed to the wall over an area symmetrically located around the pipe penetration. The Radon flux was evaluated by measuring the Radon concentration buildup in the container. The selected size of the container was the result of a theoretical analysis performed to insure that the modifications introduced in the Radon flow pattern by imposing a higher Radon concentration over a fraction of the wall surface were negligible. The analysis is reported in Appendix 5.1. After the Radon

concentrations were recorded, the water manometer was disconnected and the penetration pipe was used for recording Radon concentrations in the tailings.

5.3.2 Theoretical foundations. The diffusion coefficient of Radon through concrete (k_c) was obtained by monitoring the concentrations of Radon on both sides of a concrete wall. The measured value of the Radon flux through the wall was equated to its analytical expression which involves the diffusion coefficient, k_c and the Radon concentration on both sides of the wall (C_t and C_c). This leads to a transcendental equation in k_c , the solution of which was obtained by a numerical iteration method attributed to Newton [5-18]. The formulation of the transcendental equation results from the following reasoning:

As a result of the order-of-magnitude analyses presented in Appendix 5.1 and of adequate size selection of the Radon collecting container on the low concentration side of the concrete wall, the flow of Radon can be described by a one dimensional model

$$\frac{k_c}{\text{porc}} \frac{d^2 C(x)}{dx^2} - \lambda C(x) = 0 \quad (5.25)$$

With average concentrations existing in the tailings (C_t) and in the container (C_c) the solution of (5.25) is:

$$C(x) = \frac{1}{\sinh(rT)} [(C_c - C_t e^{-rT})e^{rx} + (C_t e^{rT} - C_c)e^{-rx}] \quad (5.26)$$

where

$$r = \sqrt{\frac{\lambda \text{porc}}{k_c}} = \sqrt{\frac{\lambda}{k_c^*}} \quad (\text{cm}^{-1})$$

T = thickness of the wall (cm)

The flux into the container (F) at $x = T$ (Fig. 5.2) is evaluated by

$$F = -k_c \left. \frac{dC(x)}{dx} \right|_{x=T} \quad (5.28)$$

which can be written as

$$F' = -k_c^* \left. \frac{dC}{dx} \right|_{x=T} \quad \text{with} \quad F' = \frac{F}{\rho \sigma r c} \quad (5.29)$$

in order to keep k_c^* as the only unknown. Using (5.26) to evaluate the derivative in (5.29) we find

$$F' = \frac{k_c^* r [C_c - C_t \cosh(rT)]}{\sinh(rT)} \quad (5.30)$$

If we define

$$y = (k_c^*)^{1/2} \quad (5.31)$$

Eq. (5.30) transforms into the following transcendental equation in y :

$$F' = \frac{\lambda^{1/2} y [C_t - C_c \cosh(\lambda^{1/2} T/y)]}{\sinh(\lambda^{1/2} T/y)} \quad (5.32)$$

and finally (5.31) reduces to

$$y = \frac{F' \cdot \sinh(c/y)}{a - b \cdot \cosh(c/y)} \quad (5.33)$$

where

$$c = \lambda^{1/2} T \quad (5.34)$$

$$a = C_s \quad (5.35)$$

$$b = C_c \quad (5.36)$$

To solve a transcendental equation, $f(y) = 0$, by Newton's method, one makes a first approximation x_0 to the solution and computes the next approximation by

$$x_{i+1} = x_i - \frac{f(x_i)}{f'(x_i)} \quad (5.37)$$

where $f'(x_1)$ is the first derivative of $f(y)$ evaluated at $y = x_1$. The successive values x_1 will converge to a root of $f(y)$. In the case of Eq. (5.32) we have

$$f(y) = a \cdot y - b \cdot y \cdot \cosh(c/y) - F' \cdot \sinh(c/y) \quad (5.38)$$

and

$$\frac{f(y)}{f'(y)} = \frac{a \cdot y - b \cdot y \cdot \cosh(c/y) - F' \cdot \sinh(c/y)}{a + \frac{b \cdot c}{y} \cdot \sinh(c/y) + \left[\frac{F' c}{y^2} - b \right] \cdot \cosh(c/y)} \quad (5.39)$$

The method converges to the root of the equation by the scheme depicted in Fig. 5.3. A plot of the function $f(y) = 0$ was first obtained to insure that there were not several roots close to the area of interest (Fig. 5.4).

A stopping criterion of the iterative procedure was set in terms of the ratio of the difference between two successive approximations and the magnitude of the last approximation.

After the root of (5.38) has been found, one obtains k_c by using (5.31) and (5.27), i.e.,

$$k_c = \text{porc} \cdot y^2 \quad (5.40)$$

A FORTRAN program which solves Eq. (5.33) by the above described procedure is presented in Appendix 5.2.

5.3.3 Experimental results. As a result of the analysis presented in Appendix 5.1 about the linearity of the time evolution of the Radon concentration in a properly sized collecting device, the flux into the container applied against the wall was obtained from

$$F = \frac{[C_f - C_i] \cdot V}{A \cdot T_s} \quad (5.41)$$

where C_f , C_i = final and initial concentration in the container (pCi/liter)

V = volume of container (liter)

A = surface area covered by the container (dm^2)

T_s = time lag between C_f and C_i .

The water manometer never showed any evidence of pressure differences across the concrete wall. The results of two experiments run on two different days are reported in Table 5.2. Since the porosity of the concrete was not known, two extreme values, 5% and 25%, had to be assumed prior to further confirmation by the application of either value in a more extensive model of the experimental facility (Section 5.4). The corresponding values of k_c for the average values, $F = 2.38 \times 10^{-5}$ pCi/ cm^2 , $C_t = 40,000$ pCi/liter, $C_c = 100$ pCi/liter and the extreme values porc = 5 percent and 25 percent are presented in Table 5.3, along with the corresponding relaxation distances computed by the relationship (5.15).

The values obtained for the diffusion coefficient k_c agree with the information found in the literature on concrete which was the most closely related to the diffusion of Radon. Schweite et al. [5.13], studying the porosity of mortars and concretes by means of $\text{O}_2\text{-N}_2$ diffusion measurements, list a value of the specific permeability:

$$\psi = \frac{D_{\text{eff}}}{D_o}$$

where D_{eff} = bulk diffusion coefficient of the $\text{N}_2\text{-O}_2$ system in concrete.

D_o = diffusion coefficient of $\text{O}_2\text{-N}_2$.

The values obtained are $\psi = 4.56 \times 10^{-4}$ and $D_0 = 2 \times 10^{-1} \text{ cm}^2/\text{s}$. These values result in $D_{\text{eff}} = 9 \times 10^{-5} \text{ cm}^2/\text{s}$. The same author reports data from Zagar [5.14] which states that, for concretes, ψ can vary from 10^{-5} to 10^{-2} . The experimental value of k_c obtained with the Kiefer concrete and the value of D_{eff} measured by Schweite are both located at the lower end of the range of possible values reported by Zagar. The explanation in the case of the Kiefer concrete, lies in the composition of the mix mentioned in paragraph 5.3.1. The formulation given amounts to a w/c ratio of 0.46 which, according to the calculation methods explained in Appendix 4.1, corresponds, at most, to a 14.4 percent capillary porosity of the cement paste. The aggregates used by the local contractor were later visually observed to be of the dense type. If the aggregates are assumed to have a 10 percent porosity, the overall porosity of the concrete amounts to 5.6 percent.

5.4 Field Application of the Linear Diffusion Theory: Diffusion through Multilayered Porous Medium

The importance of the Radon concentration profile generated by the application of a linear diffusion model to concrete and the use of the measured k_c in evaluating the increased gamma exposure due to the application of a sealant on a concrete wall required that the model be compared against more comprehensive measurements obtained experimentally. A model of the situation encountered around our experimental facility was developed in order to compare the theoretical predictions to the data obtained experimentally. The situation modeled is depicted in Fig. 5.1 and pertains to one of the basement walls, i.e., a layer of tailings located between a semi-infinite soil region and an

8 " thick concrete wall on which flux measurements were performed to determine the diffusion coefficient of Radon in concrete. The data collected at that location are the Radon concentration in the tailings close to the concrete wall (Table 5.2) and the Radon flux out of the concrete wall (Table 5.2). The values of the other parameters needed in the model were either measured (Chapter 4) or obtained from the literature. The parameters are

<u>Tailings</u>	Radium content: $[Ra] = 0.130 \times 10^{-9}$ g Radium/g tailings
	Porosity: $port = 0.36$
	Emanation power: $E = 0.2$
	Density: $\rho = 1.62$ g/cm ³
	Relaxation length: 70... 180 cm (Table 5.1)
<u>Soil</u>	Porosity $pors = 0.25$ [5.9]
	Relaxation length: 85... 150 cm (Table 5.1)
<u>Concrete</u>	Porosity: $porc = 0.05... 0.25$ [Appendix 5.1]
	Relaxation length: 8... 13 cm (Table 5.3)

The multilayered system under consideration (soil, tailings, concrete) was represented by a system of 3 diffusion equations of the type (5.12, 5.13) with the associated boundary conditions and interface matching conditions on fluxes (pCi/cm²s) and concentrations (pCi/cm³). The system solved was the following (Fig. 5.5).

In the soil:

$$\frac{d^2 C_{34}(x)}{dx^2} - \frac{\lambda \cdot pors}{k} C_{34} = 0 \quad (5.43)$$

In the tailings:

$$\frac{d^2 C_{14}}{dx^2} - \frac{\lambda \cdot port}{k_t} C_{14} + \frac{Ra \cdot \rho \cdot E \cdot \lambda \cdot 10^{12}}{k_t} = 0 \quad (5.44)$$

In the concrete:

$$\frac{d^2 C_{24}}{dx^2} - \frac{\lambda \cdot \text{porc}}{k_c} C_{24} = 0 \quad (5.45)$$

Matching conditions:

$$C_{34}(0) = C_{14}(0) \quad (5.46)$$

$$k_s \left. \frac{dC_{34}}{dx} \right|_{x=0} = k_t \left. \frac{dC_{14}}{dx} \right|_{x=0} \quad (5.47)$$

$$C_{14}(t_1) = C_{24}(t_1) \quad (5.48)$$

$$k_t \left. \frac{dC_{14}}{dx} \right|_{x=t_1} = k_c \left. \frac{dC_{24}}{dx} \right|_{x=t_1} \quad (5.49)$$

Boundary conditions:

$$C_{34}(-\infty) = 0 \quad (5.50)$$

$$C_{24}(x) = C_A = 5 \times 10^{-2} \text{ pCi/cm}^3 \quad \text{(Average indoor Radon concentration in the experimental facility).} \quad (5.51)$$

The general solutions of the system (5.43), (5.44) and (5.45), in terms of undetermined coefficients, is

$$C_{34}(x) = A_{34} \cdot e^{q_3 x} + B_{34} \cdot e^{-q_3 x} \quad (5.52)$$

$$C_{14}(x) = A_{14} \cdot \sinh q_1(x-t_1) + B_{14} \cdot \cosh q_1(x-t_1) + S'' \quad (5.53)$$

$$C_{24}(x) = A_{24} \cdot \sinh q_2(x-t_2) + B_{24} \cdot \cosh q_2(x-t_2) \quad (5.54)$$

where

$$q_1^2 = \frac{\lambda \cdot \text{port}}{k_t}, \quad q_2^2 = \frac{\lambda \cdot \text{porc}}{k_c}, \quad q_3^2 = \frac{\lambda \cdot \text{pors}}{k_s}, \quad S'' = \frac{[Ra] \cdot \rho \cdot E \cdot 10^{12}}{\text{port}} \quad (5.55)$$

After application of the boundary conditions (5.50) and (5.51), the application of the matching conditions (5.46) through (5.49) leads to a system of equations with the undetermined coefficients A_{14} , B_{14} , A_{34} , A_{24} :

$$A_{34} + A_{14} \cdot \sinh(Q_1 t_1) - B_{14} \cdot \cosh(Q_1 t_1) = S'' \quad (5.56)$$

$$B_{14} - A_{24} \cdot \sinh q_2(t_1 - t_2) = C_A \cosh q_2(t_1 - t_2) - S'' \quad (5.57)$$

$$A_{14} k_1 q_1 - A_{24} k_2 q_2 \cosh q_2(t_1 - t_2) = C_A k_2 q_2 \sinh q_2(t_1 - t_2) \quad (5.58)$$

$$A_{34} k_3 q_3 - A_{14} k_1 q_1 \cosh(q_1 t_1) + B_{14} k_1 q_1 \sinh(q_1 t_1) = 0 \quad (5.59)$$

The solution of the system (5.56) through (5.59), and the computation of the corresponding concentrations and fluxes, was obtained from the FORTRAN program reproduced in Appendix 5.3. The results of two computations involving the following values of the parameters, which were not uniquely specified above, is presented in Tables 5.4 and 5.5.

<u>Soil</u>	Relaxation distance: 138 cm
<u>Tailings</u>	Relaxation distance: 115 cm
<u>Concrete</u>	Porosity: 0.05 and 0.25

It can be verified that the values generated by inserting the parameters provided either by the literature or our own measurements closely match the experimental data of Fig. 5.2. The closer match with the experimental data obtained with $p = 5\%$ also confirms that the concrete is of the low porosity type as was expected from its composition and from the low measured value of the diffusion coefficient. It was expected from the theoretical model to generate values slightly higher than the ones recorded around the basement of the building, since

no provision was made in the one dimensional model for the Radon escaping through the top soil cover of the tailings.

5.5 Conclusions

The equation describing the diffusion of a radioactive gas in a porous medium was derived by using the concept of a bulk diffusion coefficient defined on the basis of the flux crossing a material surface area of porous medium. The necessity of including the porosity parameter in the case of sources which allow emanation of the diffusing gas into the capillary system was shown for either definition of the diffusion coefficient.

The application of a linear diffusion model to the flow of Radon through several porous media was shown to be a valid one. The measured value of the Radon diffusion coefficient in the concrete present in our experimental facility was determined to be three orders of magnitude smaller than the corresponding coefficient in soil or compacted sands. The above described model of diffusion will consequently be used to evaluate the results of the application of a sealant on a concrete wall.

6. GAMMA EXPOSURE INCREASE RESULTING FROM A SEALANT APPLICATION

The success encountered in applying a linear diffusion model to the diffusion of Radon throughout soil, tailings and concrete warrants the application of its result to the evaluation of gamma exposure in various situations. The formulation of the exposure resulting from infinite slabs containing constant or variable volume source strength will first be presented. This will demonstrate the need to derive the values of the volume radiation source strength from the Radon concentration profile and also to analyze the spectrum of gamma intensities emitted by Ra-226 and its daughters. The latter decomposition will allow the use of the linear attenuation coefficients, buildup factors and frequency factors associated with each energy of the spectrum.

6.1 Formulation of Exposure Doses.

All the exposures computed are evaluated 18" away from the concrete surface and by assuming the walls and the tailings to constitute infinite slabs.

6.1.1 Uncollided flux resulting from a shielded infinite slab with constant volume source strength. According to [6.1 p.376] and [6.2 p. 396]

$$\phi_u = \frac{S_v}{2\mu_s} [E_2(b_1) - E_2(b_3)] \quad (6.1)$$

where

ϕ_u = uncollided flux (photons/cm²-s)

S_v = source strength (photons/cm³-s)

μ_s = linear attenuation coefficient of the material containing the radiation source for the gamma energy considered. (cm⁻¹)

$$b_1 = \sum_i \mu_i t_i$$

μ_i = linear attenuation coefficient of the i^{th} shield for the gamma energy considered. (cm^{-1})

t_i = thickness of the i^{th} shield (cm)

$$b_3 = b_1 + \sum_i \mu_i t_i$$

$$E_2(x) = x \int_x^{\infty} \frac{e^{-t}}{t} dt$$

The numerical formulation of $E_2(x)$ is obtained [6.1 p. 411] by

$$E_2(x) = [e^{-x} - x E_1(x)] \quad (6.2)$$

where $E_1(x)$ is given by the following expressions depending on the range of its argument:

For $1 \leq x < 110$, the following Rand approximation is accurate to at least one part in a million:

$$E_1(x) = \frac{e^{-x}}{x} \left[\frac{a_0 + a_1 x + a_2 x^2 + x^3}{c_0 + c_1 x + c_2 x^2 + x^3} \right] \quad (6.3)$$

with	$a_0 = 0.2372905$	$c_0 = 2.4766331$
	$a_1 = 4.5307924$	$c_1 = 8.6660126$
	$a_2 = 5.1266902$	$c_2 = 6.1265272$

For $10^{-40} < x < 1$, ten terms of the following expansion yield an accuracy of at least one part in a million

$$E_1(x) = -0.57721566 - \ln x + \sum_{n=1}^{\infty} (-1)^{n-1} \frac{x^n}{n \cdot n!} \quad (6.4)$$

For $-1 < x < -10^{-40}$, the expansion

$$E_1(x) = -0.57721566 + \ln|x| + \sum_{n=1}^{\infty} \frac{|x|^n}{n \cdot n!} \quad (6.5)$$

is used. Ten terms in the sum yield an accuracy of at least one part in a million.

We never encountered in our work an argument x outside the range $-1 > x > +110$.

6.1.2 Uncollided Flux resulting from a shielded infinite slab with variable source strength. According to [6.1 p. 376] and [6.2 p. 396], if

$$S_v(x) = \sum_{i=1}^2 S_i e^{k_i x} \quad (6.6)$$

then,

$$\phi_u = \frac{1}{2\mu_s} \sum_{i=1}^2 S_i e^{k_i b_3 / \mu_s} \left[F_1(b_3, \frac{-k_i}{\mu_s}) - F_1(b_1, \frac{-k_i}{\mu_s}) \right] \quad (6.7)$$

where

$S_v(x)$ = space dependent volume source strength (photons/cm³sec)

ϕ_u, μ_s, b_1, b_3 = as in equation (6.1)

$$F_1(t, a) = \int_0^t e^{ab} E_1(b) db \quad (6.8)$$

$E_1(x)$ = as described by equations (6.3) through (6.5).

According to [6.2 p. 401] the numerical formulation of $F_1(t, a)$ is

$$F_1(t, a) = \frac{1}{a} (e^{at} E_1(t) - E_1[t(1-a)] - \ln|1-a|) \quad (6.9)$$

except for $a = 1$, when

$$F_1(t, a) = e^t E_1(t) + \ln t + 0.5772156 \quad (6.10)$$

6.1.3. Buildup factors. The flux actually reaching a given point is much larger than the one computed by the uncollided flux formulae presented so far. The increase in exposure at a given point is due to the photons which are scattered away from their original trajectories and reach the point under observation. For a monoenergetic isotropic point source the buildup (B) of the flux after a trajectory l in a homogeneous medium of macroscopic cross section μ , depends on the initial energy E_0 of the gammas and can be expressed by a two term exponential fit:

$$B(E_0, \mu) = \sum_{i=1}^2 A_i e^{\alpha_i \mu l} \quad (6.11)$$

The actual flux at a given point is then obtained by multiplying the uncollided flux by B.

For finite geometries which include shields (as in the case of our infinite slabs) the build up factor is introduced before integrating the source kernel:

$$d\phi(x) = \sum_{i=1}^2 A_i e^{\alpha_i \mu x} d\phi_u(\mu, x) \quad (6.12)$$

Since the kernel always involves a decaying exponential term of the type

$$d\phi_u(x) = dK e^{-\mu x} \quad (6.13)$$

where dK is a multiplicative constant depending on the type of differential source kernel, the multiplication operated on the source kernel

amounts to replacing the original kernel by a sum of two terms, each containing a fictitious macroscopic cross section ν_i and a corresponding weighting factor A_i :

$$d\phi(x) = A_1 d\phi_u(\nu_1, x) + A_2 d\phi_u(\nu_2, x) \quad (6.14)$$

where

$$\nu_i = \nu(1 + \alpha_i) \quad (6.15)$$

After integration of the source kernel, the actual flux resulting from a specific geometry will also correspond to a sum of two weighted terms each containing a fictitious linear attenuation coefficient.

The above method is strictly applicable to a geometry involving one type of material only. When the radiation has to travel through laminated media, an assumption has to be made with respect to the buildup factor in the various media. It is generally assumed that the buildup factor for the flux traveling along any ray from the source to the point under observation is given by the product of the point isotropic buildup factors for the segments of the path in the various media. If the ray travels the distances l_1 and l_2 respectively in two media in series, we have

$$d\phi = \left[\sum_{i=1}^2 A_{1i} e^{\alpha_{1i} \nu_1 l_1} \right] \left[\sum_{j=1}^2 A_{2j} e^{\alpha_{2j} \nu_2 l_2} \right] \cdot dK \cdot e^{-(\nu_1 l_1 + \nu_2 l_2)} \quad (6.16)$$

After rearranging, we have

$$d\phi = \sum_{i=1}^2 \sum_{j=1}^2 A_{1i} \cdot A_{2j} \cdot dK \cdot e^{-(\mu_{1i} x_1 + \mu_{2j} x_2)} \quad (6.17)$$

where

$$\mu_{1i} = \mu_1(1+\alpha_i) \quad (i = 1,2)$$

$$\mu_{2j} = \mu_2(1+\alpha_j) \quad (j = 1,2)$$

In general, it can be seen that after integration of the appropriate source kernel, the expression of the actual flux is the sum of 2^s terms where s is the number of media traversed. Each term is preceded by a combination of s weighting factors A_i and contains fictitious linear absorption coefficients, but has exactly the form of the uncollided flux.

6.2 Concentration Profiles.

All concentrations mentioned will be measured in pCi/cm^3 .

6.2.1 Radon concentration profiles in the gas phases. The Radon concentration profiles in the gas phases of the concrete and of the tailings were derived in the no-sealant case by the program BASFLUD reproduced in Appendix 5.3. For the cases with sealant, a similar system of diffusion equations was solved and a second program (BASFLUS) was written to generate the corresponding solutions. BASFLUS is reproduced in Appendix 6.1.

The Radon concentration profiles in the tailings and in the concrete respectively are generated by BASFLUO and BASFLUS under the following forms:

For the no sealant case,

$$C_{14}(x) = A_{14} \sinh q_1(x-TT) + B_{14} \cosh q_1(x-TT) + \frac{[Ra] \cdot \rho \cdot 10^{12}}{\text{port}} \quad (6.18)$$

$$C_{24}(x) = A_{24} \sinh q_2(x-TC) + C_A \cosh q_2(x-TC) \quad (6.19)$$

and for the sealant case

$$C_{15}(x) = A_{15} \sinh q_1(x-TT) + B_{15} \cosh q_1(x-TT) + \frac{[Ra] \cdot \rho \cdot 10^{12}}{\text{port}} \quad (6.20)$$

$$C_{25}(x) = B_{25} \cosh q_2(x-TC) \quad (6.21)$$

where

$$q_1 = \sqrt{\frac{\lambda \cdot \text{port}}{AK_1}} \quad , \quad q_2 = \sqrt{\frac{\lambda \cdot \text{porc}}{AK_2}}$$

TC, k_c , porc = thickness, diffusion coefficient and porosity of concrete.

TT, k_t , port = thickness, diffusion coefficient and porosity of the tailings.

Prior to evaluating the fluxes resulting from the profiles (6.18) to (6.21) the gamma exposures programs transforms them in sum of exponentials in order to match the pattern (6.6).

6.2.2 Radon concentration in the tailings solid phase. The Radon concentration in the sand particles of the tailings is assumed to be such that its activity is equal to $(1-E)$ times the activity of the Ra-226 per cm^3 of solid phase. The Radon concentration in pCi/cm^3 of solid tailings phase is therefore:

$$[C_{\text{sol}}]_{R_n} = \frac{[Ra] \cdot \rho \cdot 10^{12} (1-E)}{\text{port}} \text{ in } \text{pCi}/\text{cm}^3 \text{ of solid phase.} \quad (6.22)$$

6.2.3 Radium concentration in the tailings solid phase.

In the tailings' solid phase, the Radium concentration is

$$[C_{\text{sol}}]_{R_a} = \frac{[Ra] \cdot \rho \cdot 10^{12}}{(1-\text{port})} \text{ in } \text{pCi}/\text{cm}^3 \text{ of solid phase.} \quad (6.23)$$

6.3 Volume Source Strength of Each Gamma Energy.

The computation of the actual flux resulting from each gamma energy contained in the spectrum of Ra-226 and its daughters is based on the source strength per unit volume of the porous media, expressed in photons/ cm^3 -s. The data on concentration profiles were expressed in pCi per cm^3 of a specified phase (gas or solid). These concentrations need to be first corrected by a factor which expresses the fraction of the total unit volume which is occupied by the phase considered. Next a conversion is operated from pCi/cm^3 to photons/ cm^3 -s. Finally a frequency factor G_i is introduced for each gamma energy.

For example, if $C(x)$ represents the Radon concentration profile in the concrete, the corresponding volume source strength for the i^{th} gamma energy is

$$[S_V(x)]_i = C(x) \cdot G_i \cdot \text{porc} \cdot 3.7 \times 10^{-2} \quad \text{photons/cm}^3 \cdot \text{s.}$$

6.4 Linear Attenuation Coefficients and Buildup Factors.

The frequency factors and linear attenuation coefficients in concrete, sand and air of the various gamma energies were respectively obtained from [6.4] and [6.1]. The conversion factors from photons/cm³ to R/hr was obtained from [6.2 p. 19] for each gamma energy. All the values used in our computations are presented in Table 6.1.

The buildup coefficients for concrete were assumed to be valid for sand because of the close values of the linear attenuation coefficients in these two media. There was no buildup considered in air. The values of the buildup weighting factors and exponentials for sand and concrete were obtained from [6.2 p. 423] and the values used in our computations are presented in Table 6.2.

6.5 Theoretical Predictions of Increased Gamma Exposure for Various Cases.

Each case analyzed is described by the values used for the physical characteristics of the three media involved, by the Radon concentration profiles before and after the application of a totally efficient sealant and by the dose contribution of the various components: solid phase of the tailings, gas phase of the tailings and gas phase of the concrete. The small importance of the tailings gas phase compared to the tailings solid phase justified the omission of the contribution of the soil gas phase.

The large variability which is found in the diffusion properties of soils and concretes led us to analyze the two extreme cases of a wide spectrum of conditions which could be encountered in field applications. The first case corresponds to a low porosity concrete (5%) associated with a soil rather permeable to Radon. The concrete chosen corresponds to the type reported by Schwiete [5.13] and is also of the type found to exist in our experimental facilities. It has a relaxation distance of about 13 cm with respect to Radon diffusion. The presence of tailings between such a concrete and a soil with good permeability to Radon (Relaxation distance = 138 cm, Table 5.9) constitutes a case which results in a low fractional gamma exposure increase after the application of a sealant. The other extreme case, which consists of the association of a highly porous concrete (25%) and a soil not very permeable to Radon, results in the highest fractional increase in gamma exposure. According to the data reported by Zagar [5.14], porous concretes have relaxations lengths as high as 40 cm with respect to gaseous diffusion and according to the data reported in Table 5.9, soils with low diffusivity have relaxation lengths as low as 60 cm.

Several cases of practical importance were analyzed by assuming either of the two extreme cases to be applicable. A Radium content of 130 pCi/g tailings was assumed in every instance except in one case where the Radium content of the tailings was doubled to demonstrate the independence of the fractional exposure increase from the Radium content of the tailings. The tailings density (1.62 g/cm^3), emanation power (0.2) and relaxation distance for Radon diffusion in the tailings (115 cm) were kept constant in all cases.

The three basic cases analyzed consist of a 4" concrete slab, with average stopping power for gamma radiations, atop different thicknesses of tailings. No modifications were applied to the linear attenuation coefficients of concrete in the shielding calculations to account for the lower density of more porous concretes. Three typical tailings thicknesses under the 4" concrete slab were analyzed: 4", 9" and 24" of tailings. All exposures are evaluated 18" above the floor.

Each table of predictions compares the situations before and after the application of a sealant. The upper half of each table describes the Radon concentrations and fluxes at various locations in the three media considered (soil, tailings, concrete). The system of coordinates used is the one shown in Fig. 5.5. Negative fluxes indicate a flow of Radon in a direction opposite to the concrete. The bottom half of the tables presents a decomposition of the gamma exposure contributions from the different components of the system. The first contribution stems from the constant concentration of the Radon entrapped in the tailings particles. The second contribution arises from the Radon contribution in the gas phase of the tailings. Inspection of Eq. (6.18) reveals that this gaseous contribution can be broken down into a constant term (representative of what the Radon concentration would be in the gas phase of the tailings if it was not diffusing into the tailings and the concrete) minus the terms in "sinh" and "cosh" which represent the losses by diffusion in the adjacent media. The net dose from the Radon profile in the gas phase of the tailings results from the algebraic summation of these three terms. It can be observed in each case that the loss term is always less important in the cases with sealant and especially so in the cases involving a concrete with a high diffusion

coefficient. The three basic cases are presented in Tables 6.3 through 6.8. The situation relative to an increased Radium content of the tailings is analyzed in Table 6.9 and should be compared to the case of Table 6.6. The effect of increased wall thickness is demonstrated by comparing the case described in Tables 6.10 and 6.11 (8" concrete slab atop 2' of tailings) to the one presented in Tables 6.7 and 6.8 (4" concrete slab atop 2' of tailings). The large fractional increase (61.2%) associated with a thick concrete slab of high porosity should be appreciated in relation with the fact that a thick concrete wall results in a low exposure dose prior to the application of a sealant and that the 61.2% predicted increase in this thick porous wall case results in a final exposure dose lower than the dose observed with a 4" concrete slab prior to a sealant application.

Table 6.12 summarizes the results presented in Tables 6.3 to 6.8, 6.10 and 6.11.

6.6 Comparison Between Experimental Data and Theoretical Predictions.

A totally efficient sealant, which had been tested in our experimental facilities and is described in Chapter 7, was applied on the floor of an addition of the Pomona School in Grand Junction, Colorado. No assay of the tailings' Radium content is yet available but it is known to vary between 130 and 260 pCi/g, depending on the amount of slime fraction which was accidentally mixed with the sand fraction on the pile. Nelson, Haley, Patterson and Quirk conducted a survey of the concrete floor thickness and of the amount of tailings present under the concrete slab. The results indicate an irregular tailings distribution averaging around 9". The concrete thickness ranged from 4" to 6 1/2". A gamma

survey conducted by the Colorado Department of Health before and after the application of a sealant indicates an 18% increase in the gamma exposure. No analysis of the porosity of the concrete was made but the topsoil layers of the Grand Junction area are known to consist of clayish soils, the poor tillability of which was a major incentive for the use of tailings as a fill material.

The observed fractional increase in gamma exposure agrees well with the one predicted by Table 6.4 in the case of a configuration involving 4" of porous concrete and a clayish soil (Table 5.9). The fractional increase recorded would indicate that the concrete poured in Pomona School is rather permeable to Radon. A test bore of the floor will hopefully be made.

The results of a gamma survey conducted with an ionization chamber, after the sealment application, by Dr. K. J. Schiager are reproduced in Fig. 6.1. The numbers appearing on the map of the school rooms which were treated with a sealant are the recorded exposure doses in $\mu\text{R/hr}$. The readings range from 43 to 102 $\mu\text{R/hr}$.

6.7 Conclusions.

It is predicted within the limitations presented in this study that the fractional increase in gamma exposure resultings from the application of a sealant, designed to stop the Radon diffusion inside structures containing Uranium mill tailings around their foundation, will take values ranging from less than 1% to 60%, the higher values of the fractional increase being associated with low initial exposures and resulting in final exposures lower or equivalent to the final exposure involving a low fractional increase.

A summary of the predicted fractional gamma exposure increases resulting from the application of a sealant in each of the cases analyzed in this chapter is presented in Table 6.12.

7. DEVELOPMENT OF A RADON BARRIER

7.1 The Sealed Tanks Facility

The investigation of potential Radon barriers was first carried out on a small scale in sealed tanks. This procedure allowed a relatively rapid testing of a variety of products (approximately 20 days per product), the most promising of which was then applied in an experimental building erected on the Foothills Campus of Colorado State University to reproduce the conditions existing in structures containing Uranium mill tailings around their foundations.

7.1.1 Experimental arrangement. Radon barriers were originally tested in an arrangement depicted in Fig. 7.1. It was later modified according to the more effective design shown in Fig. 7.2.

The first experimental design involved two round stock watering tanks, one inverted over the other to form a sealed container. A compacted layer of Uranium mill tailings was placed in the lower tank and the concrete slab to be used for the tests was poured on the bed of tailings. The material to be tested as a Radon barrier was then applied to the surface of the concrete slab. The upper tank was sealed to the lower tank by means of an airtight gasket and a clamping arrangement that applied sufficient pressure to assure a gas tight seal even if the pressure differential between the interior of the tank and the outside atmosphere increased to several inches water gauge. Manometers were provided to determine the pressure differentials, both between the interior of the tank system and the outside atmosphere, and also between the upper and lower tanks. (Temperature changes did in fact produce pressure differentials between interior and exterior

atmospheres, but the pressures in upper and lower tanks remained equalized.) An automobile carburetor air filter with the open ends closed by metal plates was imbedded in the layer of tailings in the lower tank. This provided a void space of reasonable volume into which Radon diffused readily from the surrounding tailings and from which Radon concentration samples could be drawn. A sampling port in the lower tank was connected by tygon tubing to this void space. A similar sampling port was provided in the upper tank from which to draw samples for the measurement of Radon concentrations.

One sealed tank facility was initially constructed utilizing two 6 foot diameter tanks. In order to determine whether or not there was a vertical gradient of the Radon concentration in the tailings, three filtered void spaces were provided at different heights in the bed of tailings. The results obtained from the first experiments with this pair of tanks indicated that there was no vertical Radon concentration gradient in the tailings and that consequently, only one filtered void space would need to be provided in subsequent experiments. The 6 foot diameter tanks also proved to be unwieldy and 3 foot diameter tanks were used in all subsequent experiments. Six such pairs of tanks have been assembled and were used for testing of potential Radon diffusion barrier materials. It was also recognized that the number of experimental arrangements could be doubled and the testing procedures accelerated by adopting the design shown in Fig. 7.2. The top tank was replaced by a flat top, reducing the volume in which the Radon had to diffuse and reach an equilibrium concentration. The thickness of the concrete slab was also reduced to 3 in. to accelerate the diffusion process.

7.1.2 Experimental procedure. The effectiveness of a Radon barrier was determined by applying it to a cured concrete slab, sealing the arrangement and monitoring the Radon concentration in the cavities above and below the treated concrete slab. The equilibrium concentration reached in the upper cavity being directly proportional to the Radon flux across the treated slab, its ratio to the average concentration in the lower cavity was used as an indicator of the barrier effectiveness. After it became apparent that the various concrete slabs poured in the experimental tanks demonstrated important differences in their ability to allow Radon diffusion, it was decided that the effectiveness of potential Radon barriers would be determined by a two step procedure. The diffusion experiment was first performed with the bare slab and the equilibrium concentration reached in this first experiment was taken as a reference for a repeated diffusion experiment performed after the application of the potential Radon barriers.

7.2 Experimental Buildings

The Radon flux measurements described in Chapter 5 and the testing of the Radon barrier which was found to be the most promising on the basis of the sealed tank experiments were accomplished in two of the experimental structures described here.

7.2.1 Experimental buildings description. Discussion with personnel of the Colorado Department of Health and with the Grand Junction City Engineer provided the basis for the design of the experimental buildings. Three types of buildings were designed to represent the most prevalent types of construction in Grand Junction and other Uranium mill towns. These three types were:

1. A building with a full basement with poured concrete walls and floor.
2. A building with poured concrete foundations and columns supporting the floor joists (forming a crawl space beneath the main floor).
3. A building constructed on a concrete slab poured on grade.

In the interests of uniformity and reduction of construction costs it was decided that all of the structures above the foundations would be insulated wood frame construction covered on the outside with Celotex and composition siding and with the interiors finished with plaster-board. The floor plan dimensions for each building were 10 feet by 20 feet in order to simulate a single room of reasonable size in a typical home. A single door was provided for access to each building, but rough-in framing was provided for the addition of a window in each structure at a later date.

The design and construction of the buildings was begun in July 1970 and completed in January 1971. One pair of each of the three types of buildings was constructed; Uranium mill sand tailings were used under or around the foundation of one building in each pair (test buildings) while the other building in each pair was kept free from tailings (reference buildings). Each building was separated from the adjacent buildings by 10 feet with a minimum of 6 feet of undisturbed earth between any two buildings; this was done to prevent Radon emanating from the tailings under any one building from diffusing into another building. The general layout of the entire group of six buildings is shown diagrammatically in Fig. 7.3.

Arrangements were made with the Climax Uranium Mill in Grand Junction, with the approval of the Colorado Department of Health, to obtain approximately 85 tons of sand tailings to place around the experimental buildings. The tailings were hauled to the University by truck and stored in a ravine at the CSU Foothills Campus approximately one mile from the construction site of the experimental buildings. When the time came to place the tailings under and around the foundations, elaborate precautions were taken to prevent contamination of the surrounding area. The tailings were loaded onto a truck and then wet down with water and covered with plastic sheeting. Other partially completed experimental buildings at the construction site were also covered with plastic before the tailings were brought near that location. The tailings were manually shoveled from the truck into the sub-fill and back-fill locations so that the quantity and distribution of the tailings around the buildings would be known with reasonable accuracy.

For the experimental buildings with basements or crawl spaces, the excavations were carefully dug with nearly vertical walls and minimal space between the foundation and the undisturbed earth. For the test buildings of these two types, tailings were placed around the foundations so as to fill the excavation to within approximately 1 foot of the surface of the undisturbed ground. The remaining excavated space around the foundation was filled with ordinary earth. This procedure provided a rather uniform fill of tailings around the basement foundation approximately 1 foot thick at the bottom of the basement walls and thickening gradually towards the top to approximately 18 inches thick at a height of 6 1/2 feet above the base of the

basement walls. The test building with the basement, therefore, has approximately 25 yards or 40 tons of tailings around its foundation. The test building with the crawl space was back-filled in a similar manner and has approximately 10 yards or 15 tons of tailings surrounding its foundation.

For the test building of slab-on-grade construction, the space under the slab was leveled and filled with tailings to a depth of approximately 6 in. before the slab was poured. Consequently, the slab rests on approximately 4 yards or 6 tons of tailings. In each case, the quantities and distribution of tailings were designed to be representative of typical construction practices utilized in Grand Junction and other mill towns. However, the actual application of the tailings was done much more carefully around the experimental buildings than would have been the case in typical commercial construction.

7.2.2 Normalization of mean air residence time. Measurements of Radon concentrations (in pCi/liter) and Radon progeny concentrations (in WL) in the experimental buildings from January through May 1971, indicated a much larger degree of variability than had been anticipated. Reliable comparisons of concentrations among the various modules proved to be extremely difficult, if not impossible, to obtain. It became apparent that variations in wind direction and velocity were primarily responsible for the variability of Radon and Radon progeny concentrations in the buildings. The principle cause of the variability appeared to be the fact that the buildings were not completely finished inside, and there was a direct communication between outdoor and

indoor atmospheres through the cracks that existed between the concrete foundations and the exterior frame walls.

In order to eliminate, as completely as possible, the outside wind condition as a variable in the experiments, it was decided to caulk the cracks between the exterior walls and the concrete foundations of the buildings. However, to determine the effects of the caulking, measurements were made of the mean air residence times in the buildings before and after caulking. A Davis Halide Meter was borrowed from the Colorado Department of Health and Carbon tetrachloride was selected as a test aerosol. A calibration curve for CCl_4 in the range of 0 to 250 parts per million (ppm) was provided with the meter.

A quantity of CCl_4 , calculated to produce a concentration of approximately 250 ppm, was vaporized by means of a paint spray gun into the module to be tested through a sampling port penetrating the door of the module. A fan was used to mix the CCl_4 thoroughly with the air in the module. Air was subsequently drawn continuously from the sampling port through the halide meter, and measurements of the CCl_4 concentration were recorded during time periods ranging up to 4 hours.

Based upon the assumptions of uniform mixing and continual dilution, the data were analyzed for the best least squares fit to the equation:

$$[C]_t = [C]_0 e^{-t/T_a} \quad (7.1)$$

where $[C]_t$ = measured CCl_4 concentration (ppm)

$[C]_0$ = initial CCl_4 concentration (ppm)

t = elapsed time (minutes)

T_a = mean air residence time (minutes)

or: $1/T_a$ = fractional air exchange rate (min^{-1})

Experiments were conducted on June 8 and 10, 1971, in 7 of the 10 module spaces prior to caulking. After caulking, the air residence times were measured three times in each of the 10 spaces on June 18, 21 and 22, 1971. The results of all these measurements are summarized in Table 7.1. The dilution rates for the modules RS and TS which were used in testing the effectiveness of the Radon barrier are presented in Fig. 7.4. Individual data points have not been plotted since the correlation coefficients were uniformly high. The large discrepancy in slope between modules RS and TS before caulking is obvious. For comparison, the data obtained before caulking on June 8, and the data obtained on June 18, after caulking are shown in Fig. 7.4. The similarity in the dilution rates after caulking is readily apparent.

7.3 Experimental Results

7.3.1 Radon barriers testing. The data obtained from representative experiments conducted in the sealed tanks are shown in Figs. 7.5 through 7.11. In each graph, the Radon concentrations have been normalized to the best estimate of the "source term," i.e., the Radon concentration in the tailings beneath the concrete slab. In the cases reported, this source term has been taken as the highest of the concentrations measured in the lower tank during the course of the experiment.

The first graph (Fig. 7.5) is for an untreated concrete slab. For this slab, the equilibrium concentration above the slab was

approximately 20 percent of that below the slab. However, it was discovered that a very small shrinkage crack was formed around the perimeter of the slab as the concrete cured. The data obtained after this crack had been sealed are shown in Fig. 7.6. In this case, the equilibrium concentration above the slab was less than 10 percent of that in the tailings below the slab. The results obtained with two other reference bare slabs with a sealed shrinkage crack are reported in Figs. 7.7 and 7.8.

The tests performed with various products can be divided into three categories:

1. Tests performed with products available on the market. These were applied to the concrete slabs according to the instructions found on the containers.
2. A second series of experiments was performed by application of a technique developed by the Brookhaven National Laboratory (BNL) during their concrete-polymer studies. The Engineering Research Center of the Bureau of Reclamation in Denver cooperated in these experiments, and the application of the products to the experimental slabs was performed by several members of the two laboratories.
3. Upon the recommendation of Dr. H. G. Olson, Mechanical Engineering Department, Colorado State University, a third series of tests was performed by letting a Denver based commercial firm, involved in the application of waterproof cement and concrete coatings, come to CSU's experimental facility and submit some of their products to the Radon diffusion test.

The composition, methods of application, and performances of the various products tested will be described in the following three paragraphs.

7.3.1.1. Commerically avaiable products

1. AM-9 Chemical Grout (American Cyanamid Co.)

This product was considered to be a potential Radon barrier in view of the water tightness it provides to the materials it penetrates or covers. The technical specifications even mention that the gels are impermeable to gases and hydrocarbons. It consists of a mixture of two organic monomers (Acrylamide and N , N' methyleneacrilamide) and comes in the form of a dry powder. When dissolved in water, it produces a stiff gel when properly activated (Amonium persulfate, AP) and catalyzed (β -dimethylaminopropionitrile, DMAPN). Small quantities of Potassium ferricyanide (KFe) are added to control the reaction. The viscosity of the solution, which is about equal to water viscosity, depends on the ratio of AM-9 powder to water and the gel time is controlled by the pH of the solution, the temperature and the amounts of catalyst, promoter and inhibitor. By solidifying into a stiff gel, the chemical grout should seal all of the concrete capillaries it has penetrated. The polymerization is supposed to occur by chain formation followed by cross linking.

The AM-9 mixture which was used in the test was as follows:

(per 100 g of grout solution)

	(Grams)
Water	79
AM-9 Powder	10
DMAPN	0.4
KFe	0.002

The AP solution which was mixed with the above solution consisted of: (per 100 g of grout solution)

Water	10
AP	0.5

One gallon of the mixture was used to cover a 3 in. thick concrete slab with a 7 ft² area.

The Radon diffusion experiment through a treated slab led to an equilibrium concentration above the slab equal to 20 percent of the concentration below the slab.

2. Aerospray-70 (American Cyanamid Co.)

Aerospray-70 is a binder used as a polymetric soil stabilizer. It comes in the form of a milky white viscous liquid containing approximately 60 percent solids by weight. Before application it is diluted with water in proportions depending on the specific results desired. The binding and film property result from the coalescing of the polymer particles as the water is lost by evaporation or absorption in the surrounding media. One pint of Aerospray-70 was mixed with 7 pints of water to cover a 3 in. thick concrete slab with an area of 7 ft².

The diffusion experiment led to an equilibrium concentration approximately equal to 1 percent of the concentration below the treated slab.

3. Colma Fix Epoxy (Sika Chemical Corp.)

Colma Fix Epoxy comes as a two-component product (A & B) with the components having to be mixed just prior to application. The first solution contains a resin and its solvent; the second, the catalysts and promoters. After setting, the epoxy formed a

brownish brittle coating which delaminated from the concrete surface. The equilibrium concentration above the treated slab amounted to approximately 15 percent of the concentration below the slab.

7.3.1.2 Application of BNL products. Work accomplished in the last four years by BNL on concrete polymers has demonstrated the possibility of drastically reducing the water permeability of concrete by diffusing a low viscosity (< 10 cps) monomer-polymer system through the capillary structure of conventional concrete and polymerizing it in situ by radiation, chemical or thermal techniques. Parallel work by BNL has also demonstrated that aggregated can be bound by a similar polymerization to produce a strong, cementless material called polymer-concrete (PC). These two techniques appeared attractive to provide Radon barriers. It was decided not only to seal the concrete with the monomer system, but also to impregnate the Radium bearing tailings with similar compounds to seal the Radon in the sand. The results of the tailings impregnation technique will not be reported here as the present work is concerned with the outer Radon barrier solution only.

The Engineering Research Center of the Bureau of Reclamation in Denver offered its collaboration in performing the experimentation at CSU and the application of the monomer-polymer system was made on December 15, 1971.

Six identical concrete slabs (7 ft^2 , 3 in. thick) were ready to be treated; one was kept as a control (Fig. 7.7), the five others were treated and compared to the standard. As the results showed, this method did not lead to very consistent results due to the variability in the concrete of the various slabs. Indeed, some treated

slabs appeared to be more permeable to Radon than the standard slab. It was this observation which led to the decision that in the future a diffusion test would be run for reference on each slab prior to treating it. Nevertheless, the diffusion data obtained with the BNL monomers is of interest and we report it here. Each concrete slab was wire brushed and the loose particles were vacuumed off before the application of the following surface treatments to three of the five slabs:

Slab 1: Impregnation and coating of concrete slab with a Polyester-Styrene (50-50) mixture.

The composition of the mixture was the following:

- 1200 g Polyester
- 1200 g Styrene
- 6 g Co-naphthenate (.25% of total wt)
- 48 g Methyl Ethyl Ketone Peroxide (MEKP) (1% of total wt)

This mixture was soaked for 30 minutes in the concrete by applying a depression of 11 in. Hg to the bottom tank. A terminal coating of the same composition was applied as a sealant on the slab.

The data of the Radon concentration monitoring is shown in Fig. 7.9. The equilibrium concentration above the treated slab amounted to less than 0.1 percent of the concentration under the slab.

Slab 2: Impregnation of the concrete by a MMA-TMPTMA (60-40) mixture followed by a Polyester-Styrene (75-25) coating.

1300 g of the following mixture was first let to soak in the concrete for 3 hours (gel formation had taken place after 1 hour).

- 1200 g MMA (60%)
- 800 g TMPTMA (40%)
- 20 g Benzoyl Peroxide (7% of total wt)
- 20 g N, N dimethyl-p-toluidine (DMT) (1% of total wt)

Next a coat of Polyester-Styrene (75-25) was applied to seal the slab.

The results are shown in Fig. 7.9.

Slab 3: Slab coating with a Polyester resin.

A mixture of 100 g of Polyester and 20 g of MEKP was applied as a seal coating to the slab. This polyester mix showed very little penetration in the concrete and resulted in a dense continuous coating after polymerization had taken place.

The results are shown in Fig. 7.11. The equilibrium ratio of the concentrations above and below the sealed slab was approximately 1 percent.

7.3.1.3 Waterproof coating by Omnitech Industries, Inc.

After standard diffusion tests were run on two new bare concrete slabs (Figs. 7.7 and 7.8), the following treatment was applied to both slabs: Four coatings of epoxy-resins with various additives and fillers were applied at a rate of one per day. The four coatings consisted of:

1. Epoxy with solvent serving as a bonding agent
2. 100 percent solids epoxy pigmented with TiO_2
3. 100 percent clear epoxy mixed with vinyl chips
4. 100 percent solids clear epoxy

The total thickness of these four coatings amounted to approximately 0.16 inch.

The results were excellent. Neither slab showed a trace of Radon diffusion across the barrier since the Radon concentrations above the treated slab were still of the order of background after 15 days of experimentation.

7.3.2 Testing of Omnitech epoxy sealant in an experimental building. The success obtained with the product applied by Omnitech Industries led naturally to the testing of its effectiveness on a larger scale by its application on the floor of the slab on grade

building having a layer of tailings approximately 4 in. thick under the floor slab. The results of the application of a seamless epoxy floor of the composition described in the tank experiments led to an appreciable reduction of the average Radon concentration in the treated experimental module. The summary of the Radon concentrations measured in the slab on grade modules before and after the application of the seamless epoxy floor in the module with tailings is presented in Fig. 7.2. In spite of the large variabilities in the measurements and of the pronounced effect on the TS/RS ratio of the averaging time interval, the effectiveness of the coating became apparent by the reversal of the predominance in Radon concentration between the reference building and the one with the tailings.

7.3.3 Cost evaluation of Omnitech epoxy sealant. As of August 1972, coatings similar to the type described above were available from Omnitech Industries at a cost of \$1.30 per square foot plus \$1.40 per lineal foot of base. The materials and labor provided for the quoted price were described as follows:

Remove all V.A. tile, base, and mastic, properly prepare floor and seal all cracks with epoxy caulk. Integral cove base to be included in total price.

Install OmniTuff decorative quartz seamless flooring (and base) in Owner's choice of color.

Complete seamless application, after preparation, to be as follows:

First prime coat to be 60% solids water resistant epoxy primer at 3 mils thickness.

Second coat to be 100% solid, pigmented epoxy, no less than 15 mils thickness.

Third coat to be 100% solids, clear epoxy, at no less than 15 mils thickness, with 3-M Company's ceramic coated quartz granules broadcast at no less than 1/2 pound per square foot of floor.

Fourth coat to be 100% solids, clear epoxy, glaze coat at approximately 10 mils thickness.

Fifth coat to be 100% solids, clear epoxy, glaze coat at approximately 6 mils thickness.

Final texture to be smooth pebbled surface, easily cleaned.

8. CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE WORK.

The feasibility and consequences of the inner Radon barrier solution to the problem of high indoor Radon progeny concentrations has required an interdisciplinary study, the major aspects of which will be summarized here.

8.1 Conclusions on the Theoretical Aspects.

The modifications introduced by a Radon barrier in the Radon concentration profiles throughout a concrete wall and in the tailings behind it, were derived by application of a linear diffusion model to the concentration driven diffusion of a radioactive gas through a multilayered system of porous media. The resulting increase in gamma exposure was evaluated by means of shielding calculations involving infinite slabs containing a variable radiation source strength per unit volume. The buildup contributions were evaluated by assuming that the buildup factor for the flux along any ray from the source to the point under observation is given by the product of the point isotropic buildup factors for the segments of the paths in the various media. The predicted relative gamma exposure increases agree with the increases recorded to date.

The time evolution of the nuclides' population in a naturally occurring radioactive series was required in several instances. The development of a simply derivable recurrence relationship, describing the population of any nuclide in a series and suited for treatment by a digital computer, was found to be of great practical value.

In view of the importance of the concrete layer in the evaluation of Radon fluxes and gamma exposure increases, a survey of the literature on concrete was conducted. The explanations found in this study about the large variability to be expected in the properties of concrete encountered in future applications have proven to be of great assistance in interpreting the experimental data.

A survey of the literature pertinent to the diffusion of Radon in various porous media has revealed a lack of consistency both in the definition of the effective diffusion coefficient and in the resulting equation describing the diffusion of a radioactive gas in a porous medium. The treatment, by the author, of the same problem through the use of an unequivocally defined effective diffusion coefficient (based on the flux per total unit surface area of the porous medium) has led to a diffusion equation which explicitly involves the porosity of the medium. The derived diffusion equation was successfully applied to model the diffusion of Radon through a multilayered system of porous media.

A method of determination of the effective diffusion coefficient of a material from the measure of the flux resulting from imposed concentrations on both sides of a slab was developed. The method requires the solution of a transcendental equation. A Fortran program is included for the solution of the transcendental equation by Newton's algorithm.

8.2 Conclusions on the Experimental Aspects.

Repeated experiments with the Radon scintillation chambers built at very reduced cost under the supervision of Dr. K.J. Schiager have

revealed the validity of the techniques used. The discovery of Radon retention in the coatings of the scintillation chambers led to the estimate that 5% error was incurred with high concentration samples collected after insufficient flushing of the chambers.

On the basis of the theoretical considerations developed on the relationship between a concrete composition and its ability to be permeated by a gas, it was concluded that the concrete used in our experimental facility had to be of the tight variety. Confirmation of this fact was obtained by measuring its effective diffusion coefficient by the flux method mentioned above. The measured value of the diffusion coefficient was found to be in the lower range of the spectrum of values cited by Zagar [5.14] and applicable to the diffusion of gases through concrete.

Finally, the testing of several potential Radon barriers in a specially designed experimental arrangement led to the discovery of the effectiveness of a commercially available product which had been recommended to us by Dr. H.G. Olson. The barrier consists of a multi-layered coating of epoxies which demonstrate good wear properties. The effectiveness of the barrier was demonstrated by its ability to reduce the average Radon concentration in an experimental building built atop tailings to half the value observed in an identical reference structure without tailings.

8.3 Conclusions on the Economical Aspects.

A study of the potential economic impact of the Radon barrier solution was completed in January 1973 by Kukacka L.E. and Isler R.J. (BNL) [8.1] on the basis of the town of Grand Junction alone:

"Economic data have been compiled for 226 homes. Eighty homes had an EGR* < 0.001 mr/hr above background and therefore required no remedial action. For the other levels the cost estimates ranged from \$2360 to \$6560. The average for the 226 residences was \$2110. Based on the established radiation level criteria and the assumed remedial action plan, a total program cost of \$10,000,000 was estimated for the 4800 residences in Grand Junction that have tailings under or near the structure. This is $\approx 33\%$ lower than a previous estimate of \$15,000,000 which was based entirely on the removal of tailings from soil under or near areas in which the gamma radiation level was above background. Compared to the latter, the use of sealants and grouts results in an overall cost reduction of \$1,900,000 ($\approx 13\%$). The remainder of the saving (\$3,100,000) is due to the deletion of any remedial action for residences with an EGR < 0.001 mr/hr above background and for limiting tailings removal to within 5-ft of residences."

8.4 Recommendations for Future Work.

Further improvements should be incorporated in the numerical programs devised to predict gamma exposure by allowing the variable porosity of the concrete to reflect on its linear attenuation coefficient.

The importance of the concrete layer properties in the problems presented here warrant further quantitative studies of the variability of Radon diffusivities in connection with the variations imposed in its composition. The quantitative information generated by such experiments would greatly improve the quality of the predictions of gamma exposure increases due to a sealant application.

Finally, the recording of gamma exposure increases recorded with an ionization chamber after application of a sealant on the various experimental concretes just mentioned would provide the means to generate a complete theory on the interaction between a concrete composition, its diffusivity and its ability to attenuate gamma radiations.

*External gamma radiation

8.5 Applicability of the Indoor Radon Barrier Solution.

The effectiveness of an epoxy seamless coating in reducing drastically the Radon concentration in our experimental facility has demonstrated the applicability of the method in a situation free of other major pathways for the diffusion of Radon inside the treated structure. Nevertheless, Radon flux measurements conducted in the experimental basement surrounded by tailings have demonstrated that major pathways other than the concrete slabs themselves, namely the thin cracks existing at the junction of two concrete slabs, could be responsible for an appreciable portion of the Radon influx in a structure. The obtaining in field applications, of successes comparable to the ones achieved in the experimental facility, would therefore be conditioned by a concurrent careful sealing of the other preferential routes offered to the diffusion of Radon inside the treated structures.

The predictions of the gamma exposures fractional increases resulting from the application of a sealant indicate that a certain variability is to be expected depending on the tailings and concrete thicknesses and on the combination of diffusion characteristics of the soil and of the concrete involved. The survey of theoretically worst possible cases reveals that the indoor sealant method is expected to be applicable in instances of sufficiently low initial gamma exposures without exceeding the permissible level of gamma exposure. Even though the validity of the predicted gamma exposure increases seem to be confirmed by the experimental data available to date, further verifications will be necessary. Additional information is expected from controlled applications of sealants in actual structures

which were identified for their high Radon progeny level and for their relatively low gamma exposure levels.

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Table 1-1 (From [1, p. 81; 82]): Distributions of Corrected Average Gamma Levels for Locations Classified as Having Tailings.

Corrected average gamma (μ R per hour) above background	Highest average gamma for any room in structure							
	Residence average groundfloor gamma-		Business		School		Other public buildings	
	N Percent		N Percent		N Percent		N Percent	
0 ¹ -----	258	38.5	65	34.8	3	12.5	13	31.0
>0 to 10-----	278	41.5	33	17.6	5	20.8	1	2.4
10 to 20-----	70	10.4	27	14.4	2	8.3	6	14.3
20 to 30-----	37	5.5	17	9.1	1	4.2	7	16.7
30 to 40-----	11	1.6	12	6.4	2	8.3	6	14.3
40 to 50-----	4	.6	9	4.8	1	4.2	1	2.4
50 to 60-----	7	1.0	4	2.1	0	-----	4	9.5
60 to 70-----	2	.3	6	3.2	0	-----	0	-----
70 to 80-----	0	-----	4	2.1	2	8.3	2	4.8
80 to 90-----	1	.1	4	2.1	2	8.3	0	-----
90 to 100-----	0	-----	1	.5	1	4.2	0	-----
100 plus-----	2	.3	5	2.7	5	20.8	2	4.8
Total-----	670	100.0	187	100.0	24	100.0	42	100.0
Indeterminate ² -----	107	-----	7	-----	0	-----	0	-----
Total-----	777	-----	194	-----	24	-----	42	-----

¹

²Background=9 μ R per hour.

Insufficient data to permit calculation of an average.

Table 1-1. Distributions of Corrected Average Gamma Levels for Locations Classified as Having Tailings - (Continued).

DEFINITIONS AND EQUATION USED IN CALCULATING CORRECTED AVERAGE GAMMA LEVELS

CORRECTED AVERAGE GROUND FLOOR GAMMA

All gamma measurements made at floor level in rooms next to the ground and attached garages, carports, patios, breezeways, and porches are averaged and the average corrected.

CORRECTED AVERAGE GAMMA - HIGHEST ROOM

All gamma measurements at floor level in the individual room are averaged and the average corrected.

CORRECTION EQUATION (in units of $\mu\text{R/hr}$)

Corrected average gamma = .56 (uncorrected average) - 9.

Table 1.2 (From [1.1]). Integrated Air Samples Results.

<u>WL Range</u>	<u>Frequency</u>	<u>Relative Frequency (%)</u>
Control Locations:		
<0.005	9	64
0.005 - 0.013	5	36
<u>≥0.014</u>	<u>0</u>	<u>0</u>
<u>Total</u>	<u>14</u>	<u>100</u>
Tailings Locations		
<0.005	1	2
0.005 - 0.013	5	11
0.014 - 0.053	26	55
<u>≥0.054</u>	<u>15</u>	<u>32</u>
<u>Total</u>	<u>47</u>	<u>100</u>

Table 2.1. (From [1.1]) Gamma Screening Results.

<u>Period</u>	<u>Cumulative Totals</u>			
	<u>No. of Screens</u>	<u>No. With Tailings</u>	<u>No. Tailings Under</u>	<u>% Under</u>
July - Dec. 1970	3678	1983	1357	37
Jan. - Feb. 1971	5125	2801	1864	36
March - April 1971	6664	3507	2228	33
May - June 1971	8521	4108	2531	30
July - Aug. 20, 1971	10610	4853	2935	28

Table 2.2. (From [1.2]) Unit Cost Estimating Data Tabulation*

	<u>Minimum</u>	<u>Maximum</u>	<u>Average</u>	<u>Median</u>
Removal of tailings under basement slab, \$/S.F. **	\$ 2.06	\$ 7.65	\$ 4.26	\$ 4.00
Removal of tailings in crawl space, \$/C.Y. ***	22.00	125.00	42.50	26.20
Removal of tailings under slab-on-grade, \$/S.F.	1.48	11.60	4.91	4.36
Removal of tailings under carport or patio slab, or sidewalk, \$/S.F.	0.40	9.60	2.41	2.02
Removal of tailings used as backfill against foundations, \$/L.F.****	1.37	21.00	7.76	6.95
Removal of masonry fireplace containing tailings, \$/S.F.	574.00	2,388.00	1,627.81	1,522.00
Removal of masonry veneer containing tailings, \$/S.F.	3.33	9.50	4.75	3.97
Moving house from foundation to another location on property, \$/S.F.	1.54	2.08	1.81	-----
*All costs include restoration.				
**SF = Square foot				
***CY = Cubic yard				
****LF = Linear foot				

Table 3.1. Buildup of Radon Daughter's Activities in a Sealed Pure Radon Sample.

Time (m)	Rn-222 activity arbitrary unit	*Activities of daughters as fractions of Radon activity						
		Po-218	Pb-214	Bi-214	Po-214	Pb-210	Bi-210	Po-210
0	1.	0	0	0	0	0.0	0	0
15	.998	.967	.239	.0461	.0461	0.127(-7)	0.410(-11)	.381(-16)
30	.996	.999	.481	.182	.182	0.113(-6)	0.798(-10)	.160(-14)
45	.994	1.00	.649	.345	.345	0.356(-6)	0.400(-9)	.126(-13)
60	.992	1.00	.763	.497	.497	0.746(-6)	0.118(-8)	.513(-13)
75	.990	1.00	.841	.625	.625	0.127(-5)	0.261(-8)	.147(-12)
90	.989	1.00	.893	.727	.727	0.189(-5)	0.487(-8)	.339(-12)
105	.986	1.00	.929	.805	.805	0.260(-5)	0.811(-8)	.675(-12)
120	.984	1.00	.954	.863	.863	0.338(-5)	0.124(-7)	.121(-11)
135	.983	1.00	.970	.905	.905	0.420(-5)	0.179(-7)	.199(-11)
150	.981	1.00	.981	.936	.936	0.505(-5)	0.245(-7)	.310(-11)
165	.979	1.00	.989	.958	.958	0.594(-5)	0.325(-7)	.459(-11)
180	.977	1.00	.994	.973	.973	0.683(-5)	0.417(-7)	.653(-11)
195	.976	1.00	.998	.984	.984	0.775(-5)	0.522(-7)	.898(-11)
210	.974	1.00	1.00	.992	.992	0.868(-5)	0.641(-7)	.120(-10)
225	.972	1.00	1.00	.997	.997	0.961(-5)	0.772(-7)	.157(-10)
240	.970	1.00	1.00	1.00	1.00	0.105(-4)	0.918(-7)	.202(-10)
255	.968	1.00	1.00	1.00	1.00	0.115(-4)	0.108(-6)	.254(-10)
270	.966	1.00	1.00	1.00	1.00	0.124(-4)	0.125(-6)	.315(-10)
285	.964	1.00	1.00	1.00	1.00	0.134(-4)	0.144(-6)	.385(-10)
300	.963	1.00	1.00	1.00	1.00	0.143(-4)	0.164(-6)	.467(-10)

*The number between brackets represent the powers of ten by which the significant digits shown have to be multiplied.

Table 3.2. Alpha Activity Buildup in Radon Scintillation Chambers Number 36 and 44.

Total time (m)	Theoretical activity initial activity	Left counter - ChN ² 36		Right Counter - ChN ² 44	
		Counts/5m.	Multiple of init. activity	Counts/5m.	Multiple of init. activity
0	1.000	(63,571)	1.000	(34,698)	1.000
15	2.0097	127,759	2.0097	69,732	2.0098
30	2.173	140,588	2.211	82,057	2.365
45	2.332	149,945	2.358	88,858	2.561
60	2.479	158,895	2.499	92,357	2.661
75	2.601	166,660	2.622	96,844	2.791
90	2.697	171,276	2.694	99,527	2.868
105	2.768	175,711	2.764	102,115	2.942
120	2.820	178,026	2.800	103,645	2.987
135	2.856	180,190	2.834	104,000	2.997
150	2.881	180,948	2.846	104,541	3.013
165	2.897	180,829	2.844	104,316	3.006
180	2.907	180,460	2.838	104,441	3.010
195	2.912	180,068	2.832	103,550	2.984
210	2.914	179,896	2.830	102,827	2.963
225	2.913	179,955	2.831	102,436	2.952
240	2.912	179,285	2.820	101,682	2.930
255	2.909	177,443	2.791	101,289	2.919
270	2.905	176,651	2.779	100,545	2.898
285	2.901	175,483	2.760	98,642	2.843
300	2.896	175,161	2.755	98,608	2.842

Table 3.3. Theoretical Decay in Activity of an Initially Pure Sample Containing Respectively a Unit Activity of Each of the Short Lived Radon Daughters.

Time (m.)	*Activities of Radon Progeny						
	Po-218	Pb-214	Bi-214	Po-214	Pb-210	Bi-210	Po-210
0	1.00	1.00	1.00	1.00	0.00	0.00	0.00
15	0.331(-1)	0.761	0.954	0.954	0.907×10^{-6}	0.658×10^{-9}	0.115×10^{-13}
30	0.109(-2)	0.519	0.818	0.818	0.173×10^{-5}	0.256×10^{-8}	0.904×10^{-13}
45	0.362(-4)	0.352	0.656	0.656	0.240×10^{-5}	0.555×10^{-8}	0.298×10^{-12}
60	0.120(-5)	0.239	0.504	0.504	0.294×10^{-5}	0.941×10^{-8}	0.684×10^{-12}
75	0.397(-7)	0.162	0.378	0.378	0.334×10^{-5}	0.139×10^{-7}	0.129×10^{-11}
90	0.131(-8)	0.110	0.277	0.277	0.364×10^{-5}	0.189×10^{-7}	0.214×10^{-11}
105	0.435(-10)	0.746×10^{-1}	0.200	0.200	0.385×10^{-5}	0.243×10^{-7}	0.327×10^{-11}
120	0.144(-11)	0.507×10^{-1}	0.143	0.143	0.401×10^{-5}	0.299×10^{-7}	0.469×10^{-11}
135	0.477(-13)	0.344×10^{-1}	0.101	0.101	0.413×10^{-5}	0.358×10^{-7}	0.640×10^{-11}
150	0.158(-14)	0.233×10^{-1}	0.713×10^{-1}	0.713×10^{-1}	0.420×10^{-5}	0.417×10^{-7}	0.842×10^{-12}
165	0.523(-16)	0.158×10^{-1}	0.499×10^{-1}	0.499×10^{-1}	0.426×10^{-5}	0.477×10^{-7}	0.107×10^{-10}
180	0.173(-17)	0.107×10^{-1}	0.347×10^{-1}	0.347×10^{-1}	0.429×10^{-5}	0.538×10^{-7}	0.134×10^{-10}
195	0.573(-19)	0.729×10^{-2}	0.241×10^{-1}	0.241×10^{-1}	0.432×10^{-5}	0.599×10^{-7}	0.164×10^{-10}
210	0.189(-20)	0.494×10^{-2}	0.166×10^{-1}	0.166×10^{-1}	0.434×10^{-5}	0.661×10^{-7}	0.196×10^{-10}
225	0.627(-22)	0.335×10^{-2}	0.115×10^{-1}	0.115×10^{-1}	0.436×10^{-5}	0.723×10^{-7}	0.232×10^{-10}
240	0.208(-23)	0.228×10^{-2}	0.788×10^{-2}	0.788×10^{-2}	0.436×10^{-5}	0.785×10^{-7}	0.272×10^{-10}
255	0.687(-25)	0.154×10^{-2}	0.541×10^{-2}	0.541×10^{-2}	0.437×10^{-5}	0.846×10^{-7}	0.314×10^{-10}
270	0.227(-26)	0.105×10^{-2}	0.371×10^{-2}	0.371×10^{-2}	0.437×10^{-5}	0.908×10^{-7}	0.360×10^{-10}
285	0.753(-28)	0.711×10^{-3}	0.254×10^{-2}	0.254×10^{-2}	0.438×10^{-5}	0.970×10^{-7}	0.409×10^{-10}
300	0.249(-29)	0.482×10^{-3}	0.173×10^{-2}	0.173×10^{-2}	0.438×10^{-5}	0.103×10^{-6}	0.461×10^{-10}

*Same as in Table 3.1

Table 3.4. Activities of Radon Progeny After 4 hours of Buildup in an Initially Pure Ra-226 Solution of Unit Activity.

Nuclide	* Absolute activity (Arbitrary units)	
Ra-226	.999999	
Rn-222	0.297830	(-1)
Po-218	0.292448	(-1)
Pb-214	0.245032	(-1)
Bi-214	0.210255	(-1)
Po-214	0.210255	(-1)
Pb-210	0.117619	(-6)
Bi-210	0.709220	(-9)
Po-210	0.127009	(-12)

*Same as in Table 3.1.

Table 3.5 Buildup of Radium Progeny in an Initially Pure Ra-226 Sample of Unit Activity.

Days	Ra-222 Activity Arbitrary Unit	*Activities of Ra-226 as a Fraction of Parent Activity							
		Rn-222	Po-218	Pb-214	Bi-214	Po-214	Pb-210	Bi-210	Po-210
0	1.000	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
2	0.9999	0.3042	0.3039	0.3004	0.2979	0.2979	0.2717(-4)	0.2350(-5)	0.5911(-8)
4	0.9999	0.5159	0.5157	0.5133	0.5116	0.5116	0.9979(-4)	0.1676(-4)	0.8778(-7)
6	0.9999	0.6632	0.6630	0.6614	0.6602	0.6602	0.2040(-3)	0.4946(-4)	0.4007(-6)
8	0.9999	0.7657	0.7656	0.7644	0.7636	0.7636	0.3302(-3)	0.1023(-3)	0.1136(-5)
10	0.9999	0.8370	0.8369	0.8361	0.8355	0.8355	0.4718(-3)	0.1750(-3)	0.2492(-5)
12	0.9999	0.8866	0.8865	0.8860	0.8855	0.8855	0.6239(-3)	0.2657(-3)	0.4651(-5)
14	0.9999	0.9211	0.9210	0.9206	0.9204	0.9204	0.7835(-3)	0.3723(-3)	0.7772(-5)
16	0.9999	0.9451	0.9450	0.9448	0.9446	0.9446	0.9482(-3)	0.4923(-3)	0.1199(-4)
18	0.9999	0.9618	0.9618	0.9616	0.9614	0.9614	0.1116(-2)	0.6236(-3)	0.1742(-4)
20	0.9999	0.9734	0.9734	0.9732	0.9732	0.9732	0.1287(-2)	0.7642(-3)	0.2416(-4)
22	0.9999	0.9815	0.9815	0.9814	0.9813	0.9813	0.1459(-2)	0.9123(-3)	0.3227(-4)
24	0.9999	0.9871	0.9871	0.9870	0.9870	0.9870	0.1633(-2)	0.1066(-2)	0.4180(-4)
26	0.9999	0.9910	0.9910	0.9910	0.9909	0.9909	0.1807(-2)	0.1225(-2)	0.5280(-4)
28	0.9999	0.9937	0.9937	0.9937	0.9937	0.9937	0.1982(-2)	0.1388(-2)	0.6529(-4)
30	0.9999	0.9956	0.9956	0.9956	0.9956	0.9956	0.2157(-2)	0.1553(-2)	0.7930(-4)
32	0.9999	0.9969	0.9969	0.9969	0.9969	0.9969	0.2333(-2)	0.1721(-2)	0.9483(-4)
34	0.9999	0.9979	0.9979	0.9978	0.9978	0.9978	0.2509(-2)	0.1891(-2)	0.1118(-3)
36	0.9999	0.9985	0.9985	0.9985	0.9985	0.9985	0.2684(-2)	0.2063(-2)	0.1304(-3)
38	0.9999	0.9989	0.9989	0.9989	0.9989	0.9989	0.2860(-2)	0.2235(-2)	0.1506(-3)
40	0.9999	0.9993	0.9993	0.9992	0.9992	0.9992	0.3036(-2)	0.2408(-2)	0.1722(-3)
42	0.9999	0.9995	0.9995	0.9995	0.9999	0.9995	0.3212(-2)	0.2582(-2)	0.1953(-3)
44	0.9999	0.9996	0.9996	0.9996	0.9996	0.9996	0.3388(-2)	0.2757(-2)	0.2200(-3)

*Same as in Table 3.1.

Table 3.6a. Evaluation of Residual Bi-214 After the Bubbling Operation; Analysis of 0.609 MeV Photopeak.

0.609 MeV Bi-214 Photopeak

1. SPECTRA ANALYSIS

Channel 10 keV/ch	Net counts before bubbling c/40 m - channel	Net counts after bubbling c/40 m - channel
57	1640	8
58	2104	52
59	2660	68
60	3290	140
61	3380	189
62	2984	160
63	2452	83
64	1724	78
65	1264	57

2. SPECTRA INTEGRATION AND COMPARISON

Total number of channels, i	Net integral before bubbling c/40 m - i chs.	Net integral after bubbling c/40 m. - i chs.	Ratio After/Before %
3	9654	489	5.06
5	14766	640	4.33
7	18594	770	4.14
9	21498	835	3.88

Table 3.6b. Evaluation of Residual Bi-214 After the Bubbling Operation; Analysis of 1.76 MeV Photopeak.

1.76 MeV Bi-214 Photopeak

1. SPECTRA ANALYSIS

Channel 10 keV/ch	Net counts before bubbling c/40 m - channel	Net counts after bubbling c/40 m - channel
164	276	30
165	370	0
166	372	0
167	438	23
168	376	17
169	440	30
170	438	25
171	344	31
172	324	18

2. SPECTRA INTEGRATION AND COMPARISON

Total number of channels, i	Net integral before bubbling c/40 m - i chs.	Net integral after bubbling c/40 m. - i chs.	Ratio After/Before %
3	1254	70	5.58
5	2064	95	4.6
7	2778	126	4.53
9	3378	174	5.15

Table 3.7. Sampling of a 364 pCi/liter Calibrated Radon Concentration.

Scintillation chamber identification	Long term background c/5 m	Sample count c/5 m	Assumed yield	Measured concentration pCi/liter	Discrepancy from actual concentration	New yield
4	34	955	.539	354	-2.75	.524
15	97	994	.512	364	0	.512
31	63	738	.414	339	-6.87	.385
33	357	1267	.435	436	+19.78	.521

Table 3.8. Consistency of the Radon Scintillation Chamber.

Scintillat. chamber identif.	Discrepancy from the mean of all four chambers %							Average %
	Exper. 1	Exper. 2.	Exper. 3	Exper. 4	Exper. 5	Exper. 6	Exper. 7	
4	-13.6	-15.0	-14.17	-11.4	-21.6	-11.5	- 5.09	-13.19
15	+ 3.12	+ 1.16	+ 2.89	+ 1.9	+ 1.9	- 2.0	- 2.4	+ 0.938
31	- 3.62	- 2.45	- 6.35	- 6.9	+ 1.9	-10.0	- 9.1	- 5.2171
33	+14.15	+16.29	+17.62	+16.42	+17.8	+23.5	+16.89	+17.52
Level of atm sampled: pCi/l	4,000	4,000	4,000	4,000	500	350	350	----

Table 3.9. Buildup of Pb-210 and Po-210 in a Scintillation Chamber as a Function of the Sample Residence Time.

Total residence time Hours	* Activities as a fraction of Radon initial activity	
	Pb-210	Po-210
0		
2	0.33278 (-5)	0.11883 (-11)
4	0.102347(-4)	0.195718(-10)
6	0.173667(-4)	0.867255(-10)
8	0.244088(-4)	0.236062 (-9)
10	0.278905(-4)	0.351793 (-9)
12	0.381791(-4)	0.911077 (-9)
14	0.449096(-4)	0.149945 (-8)
16	0.515391(-4)	0.229542 (-8)
18	0.580690(-4)	0.332819 (-8)
20	0.645010(-4)	0.462617 (-8)
22	0.708364(-4)	0.621693 (-8)
24	0.770766(-4)	0.812725 (-8)

*Same as in Table 3.1

Table 3.10 a. Background of Instrumentation.

Time	Left counter c/5m	Right counter c/5m
3/22/73		
13:20	0	0
18:23	1,1	2,1
3/23/73		
6:35	1	2
10:17	1	0
3/24/73		
9:35	8,3	1,0
14:45	1	1
3/25/73		
12:45	1	1
3/27/73		
22:15	0	1
3/28/73		
12:30	1	0
20:25	0	0

Table 3.10b. Efficiency of Instrumentation in Counting Standard Source
Calibrated at 8500 c/m.

Time	Left Counter c/m	Right Counter c/m
3/22/73		
18:23	8671, 8671, 8632	8705, 8827, 8942
24:25	8571, 8489	8391, 8469
3/23/73		
6:35	8598, 8541	8518
10:17	8600, 8704	8224, 8145
16:22	8453, 8784	8723, 8145
23:35	--	8661, 8618
3/24/73		
9:35	8483, 8614	8612, 8636
14:45	8904, 8742, 8922	--
3/25/73		
12:45	8648, 8611	--
3/27/73		
22:15	8493, 8526	8648, 8734
3/28/73		
12:30	8498, 8469	8636, 8593
20:25	8294, 8220	8557, 8693

Table 3.11. First Short Term Background Decay Experiment.

Operation*	Date-Time		Counts c/5 m
Bkgd counting	3/23/73	23:00	135
Sampling	3/23/73	23:24	---
Counting	3/24/73	9:55	863, 811
Flushing (5 l/m.)	3/24/73	10:02-10:37	---
Counting	3/24/73	15:00	176, 170
Counting	3/25/73	12:57	148, 151

*Unless the flushing operation is mentioned, the scintillation chamber is kept sealed between operations.

Table 3.12. Second Short Term Background Decay Experiment.

Operation		Date-Time	Left counter c/5 m.		Right counter c/5 m.
Bkgd. Counting		3/22/73 8:00	342		137
Sampling		3/22/73 8:43	---		---
Flushing (Sl/m.)		3/22/73 13:30- 13:40			
		Left counter (#42)		Right counter (#44)	
Total time after flushing (m.)	Theoretical activity initial activity	c/5m	Multiple of init. activity	c/5m	Multiple of init. activity
0	2.000	(83,337)	2.000	(167,385)	2.000
15	0.987	41,127	0.987	82,605	0.987
30	0.819	34,628	0.831	69,245	0.827
45	0.656	28,527	0.685	56,909	0.680
60	0.505	22,782	0.547	45,393	0.542
75	0.378	17,709	0.425	35,760	0.439
90	0.277	13,821	0.332	28,178	0.337
105	0.203	11,128	0.267	23,245	0.278
120	0.143	9,066	0.217	19,286	0.230
135	0.101	---	---	---	---
150	0.071	6,854	0.164	14,451	0.173
165	0.050	6,283	0.151	12,955	0.155
180	0.035	5,950	0.143	12,056	0.144
195	0.024	5,704	0.137	11,641	0.139
210	0.016	5,628	0.135	11,592	0.138
225	0.011	5,639	0.135	11,142	0.133
240	0.008	5,823	0.140	11,025	0.132

Table 3.13. Third Short Term Background Decay Experiment.

Operation	Date-Time	Left counter c/5 m.		Right counter c/5 m.	
Bkgd. counting	3/22/73 19:00	350		135	
sampling	3/22/73 19:18				
Counting	3/23/73 10:40	149,700		76,200	
Flushing (5 l/m.)	3/23/73 10:53- 11:18				
Total time after flushing (m.)	Theoretical activity initial activity	Left counter (#42)		Right counter	
		Counts c/5m	Multiple of init. activity	Counts c/5m	Multiple of init. activity
0	2.000	(94,481)	2.000	(46,437)	2.000
15	0.987	----	---	----	---
30	0.819	38,690	0.819	19,016	0.819
45	0.656	31,095	0.658	15,187	0.654
60	0.505	24,780	0.524	11,747	0.506
75	0.378	19,164	0.406	9,120	0.393
90	0.277	15,524	0.329	7,545	0.325
105	0.203	----	---	----	---
120	0.143	10,479	0.222	4,931	0.212
135	0.101	9,170	0.194	4,377	0.188
150	0.071	8,293	0.175	3,930	0.169
165	0.050	7,830	0.166	3,626	0.156
180	0.035	7,176	0.152	3,546	0.153
195	0.024	7,144	0.151	3,489	0.150
210	0.017	7,128	0.151	3,309	0.142
225	0.011	7,035	0.149	3,534	0.152
240	0.008	7,040	0.149	3,452	0.149
255	0.005	----	---	----	---
270	0.004	7,501	0.159	3,699	0.159

Table 3.13. Third Short Term Background Decay Experiment - (Continued).

Total time after flushing (m.)	Theoretical activity initial activity	Left counter (#42)		Right counter (#44)	
		Counts c/5m	*Multiple of init. activity	Counts c/5 m	Multiple of init. activity
285	- 0.0025	7,202	0.152	3,775	0.162
300	0.0017	7,580	0.160	3,940	0.169
Scintillation chamber #36 was removed from counter and flushed at 5 l/m. from 16:45 to 22:30. Scintillation chamber #44 was left sealed in the counter.					
690	Negl.	366	7.7(-3)	6,038	0.260
705	Negl.	440	9.3(-3)	6,040	0.260
720	Negl.	471	9.9(-3)	6,076	0.261
Scintillation chamber #44 was flushed for 14 m. (5 l/m.) from 23:20 to 23:34					
735	Negl.	509	1.08(-2)	2,297	0.099
Both scintillation chambers were flushed from 3/23/73 23:45 to 3/24/73 10:15.					
---	----	---	354	----	136
---	----	---	368	----	131

*Same as in Table 3.1.

Table 3.14. Fourth Short Term Background Decay Experiment.

Operation	Date-Time	Left counter c/5 m.	Right counter c/5 m.
Bkgd. counting	3/27/73 22:45	335	135
Sampling	3/27/73 23:00		
Counting	3/28/73 13:00	88,206	137,701
Flushing (2 l/m.)	3/28/73 Starts 13:11		

Total time after flushing (m.)	Theoretical activity initial activity	Left counter (#36)		Right counter (#44)	
		Counts c/5m	Multiple of init. activity	Counts c/5m	Multiple of init. activity
0	2.000	(56,500)	2.000	(89,500)	2.000
15	0.987	28,123	0.995	44,200	0.988
30	0.819	22,900	0.813	36,435	0.814
45	0.656	16,219	0.574	25,312	0.565
60	0.505	13,745	0.486	21,457	0.479

Stop flushing of chamber #36					
75	0.378	10,213	0.316	15,559	0.347
90	0.277	7,868	0.278	11,146	1.233
105	0.203	6,378	0.225	8,372	0.187
120	0.143	5,033	0.178	5,932	0.132
135	0.101	4,249	0.150	4,192	0.094
150	0.071	3,642	0.129	3,027	0.068
165	0.050	3,245	0.015	2,098	0.046
180	0.035	2,988	0.106	1,448	0.032

Table 3.14. Fourth Short Term Background Decay Experiment - (Continued).

Total time after flushing (m.)	Theoretical activity initial activity	Left Counter (#36)		Right Counter (#44)	
		Counts c/5m	Multiple of init. activity	Counts c/5m	Multiple of init. activity
195	0.024	2,935	0.104	979	0.022
210	0.016	2,859	0.101	647	0.014
225	0.011	3,034	0.107	481	0.011
240	0.008	3,129	0.111	356	0.008
Interruption in counting					
315	0.005	3,587	0.127	26	6.0(-3) *
330	0.004	3,467	0.123	43	1.0(-4)
345	0.0025	3,623	0.128	10	3.0(-3)
360	0.0017	3,740	0.132	42	1.0(-4)
375	Negl.	3,727	0.132	-3	Negl.
390	Negl.	3,844	0.136	-5	Negl.
405	Negl.	3,870	0.137	5	Negl.
420	Negl.	7,783	0.275	6	Negl.
Flushing of chamber #36 for 5 minutes.					
440	Negl.	1,503	0.053		

*As in Table 3.1.

Table 4.1 Standard Source and Background Count Rates for the 1.76 MeV Photopeak (Bi-214)

	Total counts	Count time	Normalized counts per 10 m.
Standard Ra-226 source (8000 pCi)	17,967	100 m.	1,797
Background	329	40 m.	82
Net count rate	-----	-----	1,715

Table 4.2 Radium Content of Samples Based on 1.76 MeV Photopeak.

	Reading c/10 m.	Norm. read. c/10m-500g	Net Count c/10m-500g	Multiple of standard source	226Ra content pCi/500 g	226Ra content g/1000 dry metric tons
<u>Grand Junction tailings:</u>						
1	13,256	13,256	13,174	7.68	61,440	0.123
2	13,366	13,366	13,284	7.74	61,920	0.124
3	13,099	13,099	13,017	7.59	60,720	0.121
4	11,345	11,345	11,263	6.57	52,560	0.105
5	12,766	12,766	12,684	7.39	59,120	0.118
6	13,135	13,135	13,053	7.61	60,880	0.122
7	13,385	13,385	13,303	7.75	62,000	0.124
<u>Uravan tailings:</u>						
1	18,189	22,736	22,654	13.21	105,680	0.211
2	18,637	23,296	23,214	13.53	108,240	0.216
<u>Ordinary topsoil:</u>						
1	210	210	128	0.075	600	10 ⁻⁴

Table 4.3. Standard Source and Background Count Rates for the 1.46 MeV Photopeak (K-40)

	Total counts	Count time	Normalized counts per 10 m.
Standard Ra-226 source (8000 pCi)	13,492	100 m.	1,349
Background	871	40 m.	218
Net count rate	-----	-----	-----

Table 4.4. Ratios of 1.46 MeV Photopeak Activities in Samples to that in the Radium Standard.

	Reading c/10 m	Norm. read. c/10 m-500 g	Net count c/10m-500 g	Multiple of standard source
<u>Grand Junction tailings:</u>				
1	8,701	8,701	8,483	7.50
2	8,855	8,855	8,637	7.64
3	8,602	8,602	8,384	7.42
4	7,695	7,695	7,477	6.61
5	8,581	8,581	8,363	7.39
6	8,635	8,635	8,417	7.44
7	8,885	8,885	8,667	7.66
<u>Uravan tailings:</u>				
1	12,396	15,495	15,277	13.50
2	12,110	15,137	14,919	13.19
<u>Ordinary topsoil:</u>				
1	1,519	1,519	1,301	1.15

Table 4.5. Standard Source and Background Count Rates for the Integrated Spectrum from 0.5 to 2 MeV.

	Total counts	Count time	Normalized counts per 10 m.
Standard Ra-226 source (800 pCi)	517,218	100 m.	51,722
Background	27,828	40 m.	6,957
Net count rate	-----	-----	44,765

Table 4.6. Radium Content of Samples Based on Integrated Spectrum from 0.5 to 2 MeV.

	Reading c/10m.	Norm. read. c/10m-500g	Net Count c/10m-500g	Multiple of standard source	Ra-226 con- tent pCi/ 500 g	Ra-226 con- tent g/1000 dry metric tons
<u>Grand Junction tailings:</u>						
1	378,797	378,797	371,840	8.306	66,450	0.133
2	382,565	382,565	375,608	8.391	67,130	0.134
3	376,452	376,452	369,495	8.254	66,030	0.132
4	322,123	322,123	315,166	7.040	56.320	0.113
5	365,692	365,692	358,735	8.014	64.110	0.128
6	376,069	376,069	369,112	8.245	65,960	0.132
7	386,398	386,398	379,441	8.476	67.810	0.136
<u>Uravan tailings:</u>						
1	530,801	663,501	656,544	14.866	117,330	0.235
2	531,905	664,881	657,924	14.697	117,580	0.235
<u>Ordinary topsoil:</u>						
1	21,353	21,353	14,396	0.321	2,570	0.005

Table 4.7. Radium Concentrations Determined by Gamma Spectrum Analyses.

Analytical method	Grand Junction tailings (recent)	Uravan tailings (old)	Topsoil, CSU campus
Radium concentration by photopeak analysis, 1.76 MeV	120 pCi Ra-226/g (or 0.12g Ra-226 per 1000 metric tons)	214 pCi Ra-226/g (or 0.21g Ra-226 per 1000 metric tons)	1.2 pCi, Ra-226/g soil
"Radium equivalent," by spectrum integration, 0.5 to 2 MeV.	130 pCi/g (Ra-226) (or 0.13g per 1000 metric tons)	235 pCi/g (Ra-226) (or 0.23g per 1000 metric tons)	5.1 pCi/g. Ra-226/g. soil

Table 4.8. Buildup of Radium Progeny in a Ra-226 Solution.

Time (Days)	*Activities of RA-226 Daughters as Fractions of Parent Activity							
	Ra-226	Rn-222	Po-218	Pb-214	Bi-214 Po-214	Pb-210	Bi-210	Po-210
0	1.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
1	0.9999988	0.166	0.165	0.161	0.158	0.684(-5)	0.295(-6)	0.357(-9)
2	0.9999976	0.304	0.304	0.300	0.298	0.272(-4)	0.235(-5)	0.591(-8)
3	0.9999965	0.420	0.419	0.416	0.414	0.587(-4)	0.753(-5)	0.290(-7)
4	0.9999953	0.516	0.516	0.513	0.512	0.998(-4)	0.168(-4)	0.878(-7)
5	0.9999941	0.596	0.596	0.594	0.593	0.149(-3)	0.306(-4)	0.204(-6)
6	0.9999929	0.663	0.663	0.661	0.660	0.204(-3)	0.495(-4)	0.401(-6)
7	0.9999918	0.719	0.719	0.718	0.716	0.265(-3)	0.734(-3)	0.703(-6)
8	0.9999906	0.766	0.766	0.764	0.764	0.330(-3)	0.102(-3)	0.114(-5)
9	0.9999894	0.805	0.804	0.803	0.803	0.399(-3)	0.136(-3)	0.172(-5)
10	0.9999882	0.837	0.837	0.836	0.835	0.471(-3)	0.175(-3)	0.249(-5)
20	0.9999766	0.973	0.973	0.973	0.973	0.129(-2)	0.764(-3)	0.242(-4)
25	0.9999707	0.989	0.989	0.989	0.989	0.172(-2)	0.114(-2)	0.471(-4)
30	0.9999648	0.99567	0.99567	0.99565	0.99564	0.216(-2)	0.155(-2)	0.793(-4)
32	0.9999625	0.99699	0.99699	0.99698	0.99697	0.233(-2)	0.172(-2)	0.948(-4)
34	0.9999601	0.99791	0.99790	0.99789	0.99789	0.251(-2)	0.189(-2)	0.112(-3)
36	0.9999578	0.99855	0.99855	0.99854	0.99854	0.268(-2)	0.206(-2)	0.130(-3)
38	0.9999555	0.998992	0.998992	0.998987	0.998983	0.286(-2)	0.223(-2)	0.150(-3)
40	0.9999531	0.999301	0.999300	0.999297	0.999294	0.305(-2)	0.241(-2)	0.172(-3)
42	0.9999508	0.999515	0.999515	0.999513	0.999511	0.321(-2)	0.258(-2)	0.195(-3)
44	0.9999484	0.999665	0.999665	0.999663	0.999662	0.339(-2)	0.276(-2)	0.220(-3)
45	0.9999473	0.999722	0.999721	0.999720	0.999719	0.348(-2)	0.284(-2)	0.233(-3)

*Same as in Table 3.1.

Table 4.9. Density of Non-Compacted Tailings.

Sample volume and number	Total weight (g)	Net weight* (g)	Density (g/cm ³)
<u>200 cm³ samples</u>			
1	496.9	285.2	1.426
2	500.3	288.6	1.443
3	504.2	292.5	1.462
4	504.2	292.5	1.462
5	509.7	298.0	1.490
6	498.1	286.4	1.432
7	499.6	287.9	1.439
Average			<u>1.451</u>
<u>500 cm³ samples</u>			
1	943.0	731.3	1.463
2	943.5	731.8	1.463
3	941.0	729.3	1.459
Average			<u>1.462</u>
Overall average density of non-compacted tailings:			1.454

*The graduated container was weighed four times; its weight was found to be successively (in g): 211.5, 211.5, 211.9, 211.9, with an average used here of 211.7 g.

Table 4.10. Density of Compacted Tailings.

Method of compaction, sample volumes and numbers	Total weight (g)	Net weight (g)	Density (g/cm ³)
<u>Mechanical (500 cm³)</u>			
1	1021.6	809.9	1.620
2	1012.3	800.6	1.601
3	1037.6	825.9	1.652
Average			<u>1.624</u>
<u>Hydraulic (180 cm³)</u>			
1	504.2	292.5	1.625
2	509.7	298.0	1.655
3	498.1	286.4	1.591
Average			<u>1.624</u>
Overall average density of compacted tailings:			1.624

Table 4.11. Porosity of Compacted Tailings.

Experiment no.	Saturated sample*		Dry sample		Vol. of water (cm ³)	Porosity (fraction)
	Total wt. (g)	Net wt. (g)	Total wt. (g)	Net wt. (g)		
1	578.4	366.7	504.2	292.5	74.2	0.411
2	573.4	361.7	509.7	298.0	63.7	0.354
3	560.0	348.3	498.1	286.4	61.9	0.344
4	561.8	350.1	499.6	287.9	62.2	0.345
Average porosity of compacted tailings:						0.36

*The final volume of each saturated sample was 180 cm³.

Table 4.12. Tailings Gas Phase Concentrations Recorded with the Large Containers.

Date	Large container 1. C'_O (pCi/liter).	Large container 2. C'_O (pCi/liter).
1/15/73	138,407	125,082
1/18/73	132,915	121,893
1/22/73	109,088	82,249
1/24/73	128,913	107,993
1/26/73	-----	104,334
Averages	127,330	108,370

Table 4.13. Tailings Gas Phase Concentrations Recorded with the Small Containers.

Date	Large container 1. C'_O (pCi/liter).	Large container 2. C'_O (pCi/liter).
1/15/73	50,750	44,319
1/18/73	55,537	37,186
1/22/73	43,515	38,392
1/24/73	56,304	41,961
1/26/73	49,315	37,342
Averages	51,083	39,840

Table 5.1. Effective diffusion coefficients of Radon in Various Media According to Tanner [5-10] and Kraner [5-9].

Description of medium	Diffusion coeff. D cm ² /s	Relaxation distance cm
<u>AIR</u>		
Various conditions	1.0×10^{-1}	218
15° C, 76 mm Hg	1.2×10^{-1}	239
<u>SAND</u>		
• Building sand (1.4 gr/cm ³ , 39% porosity, 4% moisture)	5.4×10^{-2}	160
• Fine quartz sand (no moisture)	6.8×10^{-2}	180
• Fine quartz sand (85% moisture)	1.0×10^{-2}	69
• Fine quartz sand (15% moisture)	1.0×10^{-2}	69
• Fine sand (17% moisture)	5.0×10^{-3}	49
<u>SOILS</u>		
• Yucca flats (25% porosity)	$3.6 \times 10^{-2} *$	131
• Granodiorite	4.5×10^{-2}	146
• Metamorphic rock	1.8×10^{-2}	92
• Granite	1.5×10^{-2}	84
• Loam	8.0×10^{-3}	62
• Varved clay	7.0×10^{-3}	57

*Kraner

Table S.2. Experimental Results of Radon Flux Measurements.

C_t pCi/liter	C_i pCi/liter	C_f pCi/liter	t hr	F pCi/cm ² -s
39,152	73	231	4.29	2.6×10^{-5}
40,481	55	204	4.54	2.14×10^{-5}

Table S.3. Diffusion Coefficient and Relaxation Distance of Radon in Ordinary Concrete.

p —	k_c cm ² /s	R cm
0.05	1.69×10^{-5}	12.7
0.25	3.08×10^{-5}	7.6

Table 5.4. Theoretical Predictions of Radon Concentrations and Fluxes in a Multilayered Porous Medium.
Concrete Porosity = 5%.

CONCENTRATION DISTRIBUTIONS.

STEP SIZES.Dx1=	13.750cm	Dx2=	5.080cm	Dx3=	241.523cm
LOCATION (1) (cm)	CONCENTRATION (pCi/cm ³)		FLUX (2) (pCi/cm ² -s)		
0.	.40161E(+02)		-.29099E(-02)		
.13750E(+02)	.43622E(+02)		-.21301E(-02)		
.27500E(+02)	.46033E(+02)		-.13808E(-02)		
.41250E(+02)	.47428E(+02)		-.65119E(-03)		
.55000E(+02)	.47828E(+02)		.69079E(-04)		

CONCENTRATION DISTRIBUTION IN CONCRETE.

LOCATION (1)	CONCENTRATION	FLUX (2)
.55000E(+02)	.47828E(+02)	.69079E(-04)
.60080E(+02)	.30389E(+02)	.48493E(-04)
.65160E(+02)	.17889E(+02)	.35787E(-04)
.70240E(+02)	.82956E(+01)	.28895E(-04)
.75320E(+02)	.50000E(+01)	.26698E(-04)

CONCENTRATION DISTRIBUTION IN SOIL.

LOCATION (1)	CONCENTRATION	FLUX (2)
0.	.40161E(+02)	-.29099E(-02)
-.24152E(+03)	.69789E(+01)	-.12591E(-04)
-.48305E(+03)	.12128E(+01)	-.21880E(-05)
-.72457E(+03)	.21075E(+00)	-.38022E(-06)
-.96609E(+03)	.36622E(-01)	-.66607E(-07)

(1). Reference is made to Figure 5.4.

(2). Negative fluxes correspond to Radon flow away from the concrete.

Table 5.5. Theoretical Predictions of Radon Concentrations and Fluxes in a Multilayered Porous Medium.
Concrete Porosity = 25%, (1), (2) See Table 5.4.

CONCENTRATION DISTRIBUTIONS.

STEP SIZES, DX1= 13.750cm DX2= 5.080cm DX3= 241.523cm

CONCENTRATION DISTRIBUTION IN THE TAILINGS.

LOCATION (1) (cm)	CONCENTRATION (pCi/cm ³)	FLUX (2) (pCi/cm ² -sec)
0.	.39201E(+02)	-.28404E(-02)
.13750E(+02)	.42559E(+02)	-.20500E(-02)
.27500E(+02)	.44852E(+02)	-.12890E(-02)
.41250E(+02)	.46112E(+02)	-.54648E(-03)
.55000E(+02)	.46358E(+02)	.18825E(-03)

CONCENTRATION DISTRIBUTION IN CONCRETE.

LOCATION (1)	CONCENTRATION	FLUX (2)
.55000E(+02)	.46358E(+02)	.18825E(-03)
.60080E(+02)	.23558E(+02)	.98286E(-04)
.65160E(+02)	.11507E(+02)	.53169E(-04)
.70240E(+02)	.47050E(+01)	.32309E(-04)
.75320E(+02)	.50000E(-01)	.26191E(-04)

CONCENTRATION DISTRIBUTION IN SOIL.

LOCATION (1)	CONCENTRATION	FLUX (2)
0.	.39201E(+02)	-.28404E(-02)
-.24152E(+03)	.68120E(+01)	-.12591E(-04)
-.48305E(+03)	.11838E(+01)	-.21880E(-05)
-.72457E(+03)	.20571E(+00)	-.38022E(-06)
-.96609E(+03)	.35746E(-01)	-.66072E(-07)

Table 6.1. Frequency Factors (G_i), Conversion Factors (D_i), and Linear Attenuation Coefficients in Concrete (μ_c), Sand (μ_t), and Air (μ_a) of the Gamma Energies Emitted by Ra-226 and Its Short Lived Daughters.

No.	Nuclide	Energy MeV	$G_i \times 10^2$	$D_i \times 10^6$ $\text{Rhr}^{-1}/\text{photon cm}^2\text{s}^{-1}$	μ_c cm^{-1}	μ_t cm^{-1}	$\mu_a \times 10^3$ cm^{-1}
1	Bi-214	0.609	47	1.2	0.184	0.174	0.091
2	Pb-214	0.352	36	0.7	0.230	0.246	0.115
3	Pb-214	0.295	19	0.6	0.248	0.288	0.125
4	Bi-214	1.120	17	2.1	0.138	0.129	0.072
5	Bi-214	1.764	17	2.9	0.115	0.102	0.058
6	Bi-214	1.238	6	2.3	0.129	0.123	0.070
7	Bi-214	0.769	5	1.55	0.165	0.150	0.084
8	Bi-214	1.378	5	2.5	0.122	0.114	0.067
9	Bi-214	2.204	5	3.5	0.092	0.078	0.048
10	Pb-214	0.242	4	4.7	0.271	0.348	0.134
11	Bi-214	7.4	4	2.5	0.122	0.114	0.067
12	Pb-210	0.047	4	0.13	0.984	0.849	0.226
13	Bi-214	0.935	3	1.8	0.152	0.138	0.077
14	Bi-214	1.728	3	2.9	0.115	0.102	0.058
15	Bi-214	1.509	2	2.65	0.117	0.108	0.062
16	Bi-214	1.848	2	3.1	0.106	0.096	0.055
17	Bi-214	2.445	2	3.6	0.046	0.03	0.024
18	Pb-214	0.053	1	0.13	0.984	0.849	0.226
19	Bi-214	2.117	1	3.4	0.101	0.084	0.053
20	Rn-222	0.51	0.078	0.13	0.198	0.192	0.101
21	Po-214	0.799	0.014	1.55	0.165	0.150	0.084
22	Po-210	0.803	0.0011	0.14	0.165	0.150	0.084
23	Ra-226	0.186	4	0.33	0.420	0.299	0.150
24	Ra-226	0.26	0.007	0.52	0.264	0.315	0.132
25	Ra-226	0.42	0.0002	0.85	0.212	0.217	0.110
26	Ra-226	0.61	0.0002	1.2	0.184	0.174	0.091

Table 6.2. Build-up Coefficients for the Gamma Energies Emitted by Ra-226 and Its Short Lived Daughters.

	Energy MeV	A_1	A_2	α_1	β_2
1	0.609	11.75	-10.75	-0.104	0.014
2	0.352	13.30	-12.30	-0.125	0.002
3	0.295	13.75	-12.75	-0.131	0.0
4	1.120	9.25	- 8.25	-0.084	0.03
5	1.765	7.0	- 6.0	-0.071	0.052
6	1.238	8.8	- 7.8	-0.081	0.037
7	0.769	10.95	- 9.95	-0.097	0.020
8	1.378	8.3	- 7.3	-0.078	0.042
9	2.204	5.8	- 4.8	-0.066	0.062
10	0.242	14.1	-13.1	-0.137	0.0
11	1.4	8.2	- 7.2	-0.077	0.0425
12	0.047	15.45	-14.45	-0.16	0.0
13	0.935	10.10	- 9.10	-0.90	0.027
14	1.728	7.05	- 6.05	-0.072	0.046
15	1.509	7.80	- 6.80	-0.075	0.045
16	1.848	6.72	- 5.72	-0.07	0.054
17	2.445	5.4	- 4.4	-0.064	0.066
18	0.053	15.3	-14.3	-0.158	0.0
19	2.117	6.0	- 5.0	-0.067	0.06
20	0.51	12.4	-11.4	-0.112	0.008
21	0.799	10.8	- 9.8	-0.095	0.021
22	0.803	10.75	- 9.75	-0.094	0.022
23	0.186	14.45	-13.45	-0.0143	0.0
24	0.260	13.9	-12.9	-0.134	0.0
25	0.420	12.8	-11.8	-0.118	0.005
26	0.610	11.6	-10.6	-0.103	0.0145

Table 6.3. Gamma Exposure Fractional Increase Analysis - Case 1a.

Case 1a 4" concrete, porosity 0.05, relaxation distance 12.7 cm
 4" tailings, 130 pCi/g
 semi-infinite soil, relaxation distance 138 cm

RADON CONCENTRATIONS AND FLUXES:

Location cm	Before Sealant Application		After Sealant Application	
	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)	Concentration (pCi/cm ³)	Flux (pCi/cm ² s)
Soil:	-0.644(+3)	0.102	-0.681(-6)	0.104
	-0.322(+3)	0.105(+1)	-0.702(-5)	-0.753(-5)
	0.0	0.109(+2)	0.107(+1)	-0.777(-4)
Tailings:	0.338(+1)	0.111(+2)	0.110(+2)	-0.801(-3)
	0.677(+1)	0.112(+2)	0.112(+2)	-0.530(-3)
	0.101(+2)	0.112(+2)	0.114(+2)	-0.600(-3)
Concrete:	0.135(+2)	0.112(+2)	0.114(+2)	0.101(-4)
	0.169(+2)	0.711(+1)	0.981(+1)	0.638(-5)
	0.203(+2)	0.343(+1)	0.887(+1)	0.308(-5)
		0.500(-2)	0.169(-4)	0.856(+1)
				0.0

GAMMA EXPOSURE DOSES (μR/hr):

Components	Before Sealant Application		After Sealant Application	
Tailings particles		25.63		25.63
Tailings gas constant	6.41		6.41	
Tailings gas losses	-2.77		-2.77	
Tailings gas net	3.64	3.64	3.64	3.64
Concrete		0.11		0.30
Total		29.38		29.57
Gamma Exposure Fractional Increase: 1.66%				

Table 6.4. Gamma Exposure Fractional Increase Analysis - Case 1b.

Case 1b 4" concrete, porosity 0.25, relaxation distance 43.6 cm
 4" tailings, 130 pCi/g
 semi-infinite soil, relaxation distance 63.2 cm

RADON CONCENTRATIONS AND FLUXES:

Location cm	Before Sealant Application		After Sealant Application	
	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)
Soil:	-0.295(+3)	0.613(-1)	-0.312(-6)	0.182
	-0.147(+3)	0.632	-0.321(-5)	0.188(+1)
	0.0	0.652(+1)	-0.216(-3)	0.194(+2)
Tailings:	0.338(+1)	0.655(+1)	0.661(-4)	0.195(+2)
	0.677(+1)	0.648(+1)	0.349(-3)	0.196(+2)
	0.101(+2)	0.631(+1)	0.632(-3)	0.196(+2)
Concrete:	0.135(+2)	0.419(+1)	0.622(-3)	0.194(+2)
	0.169(+2)	0.209(+1)	0.617(-3)	0.192(+2)
	0.203(+2)	0.500(-2)	0.615(-3)	0.191(+2)

GAMMA EXPOSURE DOSES (μR/hr):

Components	Before Sealant Application		After Sealant Application	
Tailings particles		25.63		25.63
Tailings gas constant	6.41		6.41	
Tailings gas losses	-2.84		-2.53	
Tailings gas net	3.57	3.57	3.88	3.88
Concrete		0.33		3.22
Total		29.53		32.73
Gamma Exposure Fractional Increase: 11.17%				

Table 6.5. Gamma Exposure Fractional Increase Analysis - Case 2a.

Case 2a 4" concrete, porosity 0.05, relaxation distance 12.7 cm
 9" tailings, 130 pCi/g
 semi-infinite soil, relaxation distance 138 cm

RADON CONCENTRATIONS AND FLUXES:

Location cm	Before Sealant Application		After Sealant Application	
	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)
Soil:	-0.644(+3)	0.204	-0.681(-6)	0.207
	-0.322(+3)	0.211(+1)	-0.702(-5)	0.213(+1)
	0.0	0.217(+2)	-0.157(-2)	0.220(+2)
Tailings:	0.762(+1)	0.227(+2)	-0.103(-2)	0.230(+3)
	0.152(+2)	0.233(+2)	-0.493(-3)	0.236(+2)
	0.228(+2)	0.235(+2)	0.453(-4)	0.238(+2)
Concrete:	0.262(+2)	0.147(+2)	0.385(-4)	0.202(+2)
	0.296(+2)	0.712(+1)	0.346(-4)	0.182(+2)
	0.330(+2)	0.500(-2)	0.334(-4)	0.175(+2)

GAMMA EXPOSURE DOSES (μR/hr):

Components	Before Sealant Application		After Sealant Application	
Tailings particles		35.99		35.99
Tailings gas constant	8.99		8.99	
Tailings gas losses	-4.14		-4.12	
Tailings gas net	4.85	4.85	4.87	4.87
Concrete		0.23		0.62
Total		41.07		41.48
Gamma Exposure Fractional Increase: 0.85%				

Table 6.6. Gamma Exposure Fractional Increase Analysis - Case 2b.

Case 2b 4" concrete, porosity 0.25, relaxation distance 43.6 cm
 9" tailings, 130 pCi/g
 semi-infinite soil, relaxation distance 63.2 cm

RADON CONCENTRATIONS AND FLUXES:

Location cm	Before Sealant Application		After Sealant Application	
	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)
Soil:	-0.295(+3)	0.132	-0.312(-6)	0.337
	-0.147(+3)	0.136(+1)	-0.321(-5)	0.348(+1)
	0.0	0.140(+2)	-0.467(-3)	0.359(+2)
Tailings:	0.762(+1)	0.142(+2)	0.124(-3)	0.336(+2)
	0.152(+2)	0.138(+2)	0.717(-3)	0.370(+2)
	0.228(+2)	0.131(+2)	0.131(-2)	0.370(+2)
Concrete:	0.262(+2)	0.870(+1)	0.129(-2)	0.365(+2)
	0.296(+2)	0.434(+1)	0.128(-2)	0.361(+2)
	0.330(+2)	0.500(-2)	0.127(-2)	0.360(+2)

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GAMMA EXPOSURE DOSES (μR/hr):

Components	Before Sealant Application		After Sealant Application	
Tailings particles		35.99		35.99
Tailings gas constant	8.99		8.99	
tailings gas losses	-4.35		-3.50	
Tailings gas net	4.64	4.64	5.49	5.49
Concrete		0.69		6.06
Total		41.32		47.54
Gamma Exposure Fractional Increase: 15.05%				

Table 6.7. Gamma Exposure Fractional Increase Analysis - Case 3a.

Case 3a 4" concrete, porosity 0.05, relaxation distance 12.7 cm
 2' tailings, 130 pCi/g
 semi-infinite soil, relaxation distance 138 cm

RADON CONCENTRATIONS AND FLUXES:

Location cm	Before Sealant Application		After Sealant Application	
	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)
Soil:	-0.644(+3)	0.394	-0.681(-6)	0.398
	-0.322(+3)	0.406(+1)	-0.702(-5)	-0.288(-4)
	0.0	0.419(+2)	0.410(+1)	-0.297(-3)
Tailings:	0.200(+2)	-0.303(-2)	0.423(+2)	-0.306(-2)
	0.400(+2)	-0.194(-2)	0.473(+2)	-0.198(-2)
	0.497(+2)	-0.907(-3)	0.503(+2)	-0.953(-3)
Concrete:	0.600(+2)	0.505(+2)	0.512(+2)	0.453(-4)
	0.633(+2)	0.318(+2)	0.438(+2)	0.285(-4)
	0.677(+2)	0.153(+2)	0.396(+2)	0.137(-4)
	0.701(+2)	0.500(-2)	0.382(+2)	0.0

GAMMA EXPOSURE DOSES (μ R/hr):

Components	Before Sealant Application		After Sealant Application	
Tailings particles		40.90		40.90
Tailings gas constant	10.22		10.22	
Tailings gas losses	-3.43		-3.41	
Tailings gas net	6.79	6.79	6.81	6.81
Concrete		0.51		1.35
Total		48.20		49.06
Gamma Exposure Fractional Increase: 1.78%				

Table 6.8. Gamma Exposure Fractional Increase Analysis - Case 3b.

Case 3b 4" concrete, porosity 0.25, relaxation distance 43.6 cm
 2' tailings, 130 pCi/g
 semi-infinite soil, relaxation distance 63.2 cm

RADON CONCENTRATIONS AND FLUXES

Location cm	Before Sealant Application		After Sealant Application	
	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)
Soil:	-0.295(+3)	0.306	-0.312(-6)	---
	-0.147(+3)	0.316(+1)	-0.321(-5)	---
	0.0	0.326(+2)	-0.108(-2)	---
Tailings:	0.200(+2)	0.335(+2)	0.183(-3)	0.642(+2)
	0.400(+2)	0.318(+2)	0.145(-2)	-0.120(-2)
	0.600(+2)	0.276(+2)	0.658(+2)	-0.424(-3)
Concrete:	0.633(+2)	0.277(-2)	0.659(+2)	0.345(-3)
	0.677(+2)	0.183(+2)	0.650(+2)	0.229(-3)
	0.701(+2)	0.914(+1)	0.644(+2)	0.114(-3)
		0.500(-2)	0.643(+2)	0.0

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GAMMA EXPOSURE DOSES (μR/hr):

Components	Before Sealant Application	After Sealant Application
Tailings particles	40.90	40.90
Tailings gas constant	10.22	10.22
Tailings gas losses	-3.19	-2.51
Tailings gas net	7.04	7.71
Concrete	1.45	10.77
Total	49.39	59.38
Gamma Exposure Fractional Increase: 20.22%		

Table 6.9. Gamma Exposure Fractional Increase Analysis - Case 4.

Case 4 4" concrete, porosity 0.25, relaxation distance 43.6 cm
 9" tailings, 260 pCi/g
 semi-infinite soil, relaxation distance 63.2 cm

RADON CONCENTRATIONS AND FLUXES:

Location cm	Before Sealant Application		After Sealant Application	
	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)
Soil:	-0.295(+3)	0.264	-0.312(-6)	0.675
	-0.147(+3)	0.273(+1)	-0.321(-5)	0.696(+1)
	0.0	0.281(+2)	-0.934(-3)	0.718(+2)
Tailings:	0.762(+1)	0.284(+2)	0.249(-3)	0.733(+2)
	0.152(+2)	0.277(+2)	0.143(-2)	0.740(+2)
	0.228(+2)	0.262(+2)	0.262(-2)	0.741(+2)
Concrete:	0.262(+2)	0.174(+2)	0.258(-2)	0.730(+2)
	0.296(+2)	0.867(+1)	0.256(-2)	0.723(+2)
	0.330(+2)	0.500(-2)	0.255(-2)	0.721(+2)

GAMMA EXPOSURE DOSES (μR/hr):

Components	Before Sealant Application		After Sealant Application	
Tailings particles		72.0		72.0
Tailings gas constant	18.00		18.00	
Tailings gas losses	-8.71		-7.02	
Tailings gas net	9.29	9.29	10.98	10.98
Concrete		1.38		12.12
Total		82.67		95.10
Gamma Exposure Fractional Increase: 15.03%				

Table 6.10. Gamma Exposure Fractional Increase Analysis - Case 5a.

Case 5a 8" concrete, porosity 0.05, relaxation distance 12.7 cm
 2' tailings, 130 pCi/g
 semi-infinite soil, relaxation distance 138 cm

RADON CONCENTRATIONS AND FLUXES:

Location cm	Before Sealant Application		After Sealant Application	
	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)
Soil:	-0.644(+3)	0.396	-0.681(-6)	0.397
	-0.322(+3)	0.408(+1)	-0.702(-5)	0.409(+1)
	0.0	0.421(+2)	-0.305(-2)	0.422(+2)
Tailings:	0.200(+2)	0.471(+2)	-0.196(-2)	0.472(+2)
	0.400(+2)	0.500(+2)	-0.930(-3)	0.501(+2)
	0.600(+2)	0.508(+2)	0.735(-4)	0.510(+2)
Concrete:	0.667(+2)	0.274(+2)	0.463(-4)	0.321(+2)
	0.735(+2)	0.119(+2)	0.326(-4)	0.226(+2)
	0.803(+2)	0.500(-2)	0.284(-4)	0.197(+2)

GAMMA EXPOSURE DOSES (μR/hr):

Components	Before Sealant Application		After Sealant Application	
Tailings particles		12.58		12.58
Tailings gas constant	3.15		3.15	
Tailings gas losses	-1.03		-1.03	
Tailings gas net	2.12	2.12	2.12	2.12
Concrete		0.36		0.83
Total		15.06		15.58
Gamma Exposure Fractional Increase: 3.45%				

Table 6.11. Gamma Exposure Fractional Increase Analysis - Case 5b.

Case 5b 8" concrete, porosity 0.25, relaxation distance 43.6 cm
 2' tailings, 130 pCi/g
 semi-infinite soil, relaxation distance 63.2 cm

RADON CONCENTRATIONS AND FLUXES:

Location cm	Before Sealant Application		After Sealant Application	
	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)	Concentration (pCi/cm ³)	Flux (pCi/cm ² -s)
Soil:	-0.295(+3)	0.385	-0.312(-6)	0.544
	-0.147(+3)	0.397(+1)	-0.321(-5)	0.561(+1)
	0.0	0.410(+2)	-0.136(-2)	0.578(+2)
Tailings:	0.200(+2)	0.425(+2)	-0.227(-3)	0.608(+2)
	0.400(+2)	0.419(+2)	0.899(-3)	0.621(+2)
	0.600(+2)	0.389(+2)	0.205(-2)	0.617(+2)
Concrete:	0.667(+2)	0.254(+2)	0.194(-2)	0.582(+2)
	0.735(+2)	0.125(+2)	0.187(-2)	0.562(+2)
	0.803(+2)	0.500(-2)	0.185(-2)	0.555(+2)

GAMMA EXPOSURE DOSES (μR/hr):

Components	Before Sealant Application		After Sealant Application	
Tailings particles		12.58		12.58
Tailings gas constant	3.15		3.15	
Tailings gas losses	-0.96		-0.79	
Tailings gas net	2.19	2.19	3.07	3.07
Concrete		1.70		10.90
Total		16.47		26.55
Gamma Exposure Fractional Increase: 61.2%				

Table 6.12 Summary of Extreme Theoretical Fractional Gamma Exposure Increases Resulting From a Sealant Application.

Tailings Thickness	4" concrete slab		8" concrete slab	
	Minimum %	Maximum %	Minimum %	Maximum %
4"	1.66	11.17	--	--
9"	0.85	15.05	--	--
24"	1.78	20.22	3.45	61.2

Table 7.1 Summary of Natural Ventilation Rate Measurements in Experimental Buildings.

	Bldg. Code	CCl ₄ Conc. (ppm) at start	Fractional Air Exchange per minute	Mean Residence Time (min.)	Correlation Coefficient
6/8/71 - Before Caulking	RS	414	-.0642	16	-.91
	TS	253	-.0061	164	-.77
	TC	198	-.0109	198	-.90
6/10/71 "	RBU	234	-.0047	213	-.96
	RBD	187	-.0061	164	-.96
	TBU	200	-.0070	143	-.99
	TBD	167	-.0060	167	-.97
Mean = 137; Std. Dev. = 64					
6/18/71 - After Caulking	RS	225	-.0039	256	-.93
	TS	236	-.0043	233	-.93
	RCU	231	-.0036	278	-.95
	RCD	190	-.0094	106	-.99
	TCU	280	-.0046	217	-.92
	TCD	264	-.0056	179	-.94
	RBU	244	-.0041	245	-.94
	RBD	197	-.0032	312	-.91
	TBU	258	-.0041	244	-.93
	TBD	219	-.0044	227	-.92
Mean = 230; Std. Dev. = 56					

Table 7.2 Summary of Radon Concentrations Measured in Slab-on-grade Modules Before and After Application of a Seamless Epoxy Floor. (K.J. Schiager, Personal Communication).

Periods	Radon concentration in module RS pCi/liter	Radon concentration in module TS pCi/liter	Ratio of concentrations TS/RS
4/27 - 6/26/71	8.5±16.0 (7)*	14.0±15.5 (7)	1.65
6/27 - 28/71	{All modules were caulked to minimize air exchange to the outside and to provide uniform mean air residence times of approx. 3 hours after caulking.}		
6/28 - 7/2/71	1.1±1.0 (5)	12.7±20.7 (5)	12.0
6/28 - 8/30/71	2.3±2.5 (24)	11.1±23.6 (24)	4.8
8/3 - 10/27/71	1.9±2.1 (23)	5.6±18.3 (28)	2.9
9/1 - 12/3/71	1.7±2.0 (16)	2.4±3.6 (20)	1.4
6/19 - 23/72	{Seamless epoxy floor installed by Omni-Tech Industries in module TS only:}		
6/26 - 8/13/72	1.9±3.2 (28)	1.0±3.8 (28)	0.53

*The Radon concentrations are expressed as averages in pCi/liter ± one Standard Derivation, with the number of measurements in parentheses.

Figure 1.1. Decay Scheme of Radium Progeny.

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Figure 1.2. Steps in Uranium and Vanadium Ore Processing
(Climax Uranium Company)

Figure 1.3. Major Exposure Pathway from Tailing Radiations.

Figure 2.1 Examples of tailings locations.

The complete solution for the case of a primary substance P and three products Q, R, S is

$$\begin{aligned}
 P &= P_0 e^{-\lambda_1 t}, \quad Q = \frac{\lambda_1}{\lambda_2 - \lambda_1} P_0 e^{-\lambda_1 t} + \frac{\lambda_1 P_0}{\lambda_1 - \lambda_2} + Q_0 e^{-\lambda_2 t} \\
 R &= \frac{\lambda_1 \lambda_2 P_0}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} e^{-\lambda_1 t} + \left[\frac{\lambda_1 \lambda_2 P_0}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{\lambda_2 Q_0}{\lambda_3 - \lambda_2} \right] e^{-\lambda_2 t} \\
 &\quad + \left[\frac{\lambda_1 \lambda_2 P_0}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} + \frac{\lambda_2 Q_0}{\lambda_2 - \lambda_3} + R_0 \right] e^{-\lambda_3 t}, \\
 S &= \frac{\lambda_1 \lambda_2 \lambda_3 P_0}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_1)} e^{-\lambda_1 t} \\
 &\quad + \left[\frac{\lambda_1 \lambda_2 \lambda_3 P_0}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)} + \frac{\lambda_2 \lambda_3 Q_0}{(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)} \right] e^{-\lambda_2 t} \\
 &\quad + \left[\frac{\lambda_1 \lambda_2 \lambda_3 P_0}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3)} + \frac{\lambda_2 \lambda_3 Q_0}{(\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3)} \right. \\
 &\quad \left. + \frac{\lambda_2 R_0}{\lambda_4 - \lambda_3} \right] e^{-\lambda_3 t} + \left[\frac{\lambda_1 \lambda_2 \lambda_3 P_0}{(\lambda_1 - \lambda_4)(\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4)} \right. \\
 &\quad \left. + \frac{\lambda_2 \lambda_3 Q_0}{(\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4)} + \frac{\lambda_3 R_0}{\lambda_3 - \lambda_4} + S_0 \right] e^{-\lambda_4 t}.
 \end{aligned}$$

The solution may evidently be obtained by superposing the solutions of the case in which the initial values of P, Q, R, S are given by

- (1) $P(0) = P_0, Q(0) = 0, R(0) = 0, S(0) = 0,$
- (2) $P(0) = 0, Q(0) = Q_0, R(0) = 0, S(0) = 0.$
- (3) $P(0) = 0, Q(0) = 0, R(0) = R_0, S(0) = 0.$
- (4) $P(0) = 0, Q(0) = 0, R(0) = 0, S(0) = S_0.$

The method is perfectly general, and the corresponding formulae for the case of $n - 1$ products may be written down at once.

Note: P_0, Q_0, R_0, S_0 are the initial populations of the respective nuclides P, Q, R, S .

Figure 3.1 Bateman Solution for the Activities of the Nuclides in a Radioactive Chain.

Figure 3.2. Radon Progeny Buildup to Secular Equilibrium.

Figure 3.3. Radon Progeny Decay in Absence of Parent Ra-222.

Figure 4.1. Determination of Tailings Emanation Power.
Experimental Arrangement.

Fig. 5.1. Basement of Experimental Building.

Fig 5.2. Measure of Radon Flux. Experimental Arrangement.

Fig 5.3. Convergence of Newton's Method for the Solution of Transcendental Equations.

Fig 5.4. Trend of Transcendental Equation Leading to a Solution for the Diffusion Coefficient.

Fig 5.5 Multilayered System of Porous Media.

Fig 6.1. Gamma Survey of Pomona School (South Building).
 After application of a sealant. Readings in
 $\mu\text{R/hr}$, 18" above floor.
 9-8-1972 (K. J. Schiager).

Legend: a. Round, stock watering tank, 3 ft. dia. x 2 ft. deep.
 b. Identical tank, inverted. c. Airtight gasket.
 d. Clamping frame. e. Uranium mill sand tailings, compacted. f. Filtered void space (auto carburetor air filter), from which to sample radon and measure gas pressure. g. Concrete test slab. h. Sealant test material applied to upper slab surface. i. Radon sampling ports. j. Manometer for measuring pressure differential between tank interior and outside atmosphere. k. Manometer for measuring pressure differential between upper and lower tanks.

Fig 7.1. First Design of Experimental Arrangement for the Testing of Radon Diffusion Barriers.

Fig 7.2. Second Design of Experimental Arrangement for the Testing of Radon Diffusion Barriers.

Fig 7.3. Perspective Diagram of the Six Experimental Buildings Used for Radon Diffusion Barrier Testing.

Fig 7.4. Dilution of CCl_4 with Time in Experimental Modules -
RS (Reference Slab on Grade) and TS (Slab on Grade with Tailings)
Before and After Caulking.

Fig 7.5. Relative Radon Concentrations in Sealed Tanks Below and Above a Concrete Slab - Untreated Slab with Unsealed Shrinkage Crack.

Fig 7.6. Relative Radon Concentrations in Sealed Tanks Below and Above a Concrete Slab - Untreated, but with Sealed Shrinkage Crack.

Fig 7.7 Relative Radon Concentrations in Sealed Tanks Below and Above a Concrete Slab - Reference Slab for BNL Tests and for Omnitech Tests.

Fig 7.8. Relative Radon Concentrations in Sealed Tanks Below and Above Bare Concrete Slab - Reference Slab 2 for Omnitech Tests.

Fig 7.9. Relative Radon Concentrations in Sealed Tanks Below and Above Concrete Slab 1, Sealed with Polyester-Styrene (BML).

Fig 7.10. Relative Radon Concentrations in Sealed Tanks Below and Above Concrete Slab 2, Sealed with Polyester-Styrene (BNL).

Fig 7.11. Relative Radon Concentrations in Sealed Tanks Below and Above Concrete Slab 3, Sealed with Polyester Resin (BNL).

APPENDIX 1.1 - RADIOACTIVE DECAY

The spontaneous radioactive disintegration of nuclides is a statistical phenomenon in the sense that one cannot predict when a specific nucleus will decay. Yet, if observations are performed a large number of times with the same species of nuclide (Ra-226 for example) one can compute an average lifetime τ which is a characteristic of the considered nuclide. If instead of repeating the experiment a large number of times with one nuclide at a time in order to establish an average lifetime, one considers simultaneously a large number N of nuclides, the same statistical regularity of the radioactive decay expresses itself in the fact that a constant fraction λ of the number of nuclides present at any time t will decay per unit time. λ is called the decay constant of the nuclide and it is used to describe our observations by the fundamental law of radioactive decay:

$$\frac{dN(t)}{dt} = -\lambda N(t) \quad (1)$$

where $N(t)$ represents the number of nuclides present at any time t

$\lambda N(t)$ represents the number of atoms which decay per unit time; it is called the activity of the population $N(t)$ and is measured in Curies. 1 Curie corresponds to 3.7×10^{10} disintegrations per second.

The third parameter, besides τ and λ , used to characterize the decay rate of a radioactive nuclide is the half-life: $T_{1/2}$; it is the time required for half of the $N(t)$ nuclides present at any time t to decay. The relationship between the three parameters τ , λ and $T_{1/2}$ is:

$$T_{1/2} = \frac{\ln 2}{\lambda} = \tau \cdot \ln 2 \quad (2)$$

APPENDIX 1.2 - SECULAR EQUILIBRIUM

Reference will be made several times to the secular equilibrium condition. The following explains its origin and meaning. In the case of a radioactive chain as depicted by Fig. 1.1, the evaluation of the number of nuclei of each kind as a function of time requires the solution of a system of first order ordinary differential equations; each equation expresses that in a radioactive chain of the type

nuclei of each intermediate kind are constantly created because of the decay of their parent but are also constantly disappearing because of their own radioactive decay. For the population $N_B(t)$ for example, the balance equation is:

$$\frac{dN_B(t)}{dt} = \lambda_A N_A(t) - \lambda_B N_B(t) \quad (2)$$

There is such an equation for each nuclide population in the chain. Harry Bateman, in 1910, solved the system formed by equations of the type (2). In our study it is realistic to assume that the Radon-222 concentration is maintained constant by the continuous decay of Radium-226 whose population barely changes in time because of its very long half-life (1602 years). In the case of a constant population of the parent A , N_{A_0} , i.e., of a constant activity of the parent $\lambda_A N_{A_0}$, the activities $I_i(t) = \lambda_i N_i(t)$ of the progeny are described by the following equations in terms of the activity of the parent A :

$$I_A(t) = \lambda_A N_{A_0} = \text{constant} \quad (3)$$

$$I_B(t) = \lambda_A N_{A_0} [1 - e^{-\lambda_B t}] \quad (4)$$

$$I_C(t) = \lambda_A N_{A_0} \left[1 - \frac{\lambda_C}{\lambda_C - \lambda_B} e^{-\lambda_B t} - \frac{\lambda_B}{\lambda_B - \lambda_C} e^{-\lambda_C t} \right] \quad (5)$$

$$I_D(t) = \lambda_A N_{A_0} \left[1 - \frac{\lambda_C \lambda_D}{(\lambda_C - \lambda_D)(\lambda_D - \lambda_B)} e^{-\lambda_B t} - \frac{\lambda_B \lambda_D}{(\lambda_B - \lambda_C)(\lambda_D - \lambda_C)} e^{-\lambda_C t} - \frac{\lambda_B \lambda_C}{(\lambda_B - \lambda_D)(\lambda_C - \lambda_D)} e^{-\lambda_D t} \right] \quad (6)$$

Equations (3) through (6) show that the activities I_B , I_C , I_D of the daughters approach the activity of the parent A as the decaying exponentials tend to zero. This situation is called secular equilibrium.

Appendix 3.1. Program DECSEK. Recurrence Solution to Bateman's Equations.

```

PROGRAM DECSEK(INPUT,OUTPUT,TAPES=(INPUT,1)APE6=OUTPUT)
DIMENSION T(10),A0(10),LAM(10),B(10,10),C(10),A(10)
REAL LAM
C PROGRAM TO EVALUATE THE ACTIVITIES OF NUCLIDES IN A RADIOACTIVE CHAIN.
C MEANING, AND UNITS OF INPUT DATA
C N=NUMBER OF NUCLIDES IN THE CHAIN
C M=NUMBER OF POINTS WHICH ARE ANALYSED IN TIME,INCLUDING T=0
C TMIN=TIME AT WHICH FIRST POINT IS COMPUTED(SAME TIME UNITS AS THE T(I))
C TMAX=TIME AT WHICH LAST POINT IS COMPUTED(SAME TIME UNITS AS THE T(I))
C TII=HALF LIVES OF THE NUCLIDES(TIME UNIT MOST APPROPRIATE TO THE PROBLEM)
C K1,K2,K3=INTEGRALS, CODE NUMBERS
C A0(I)=INITIAL ACTIVITIES OF ALL THE NUCLIDES(ONLY IF K2=3)
C DATA INPUT
C FIRST CARD      N,M,TMIN,TMAX      FORMAT 2(10,2E10,3
C NEXT N CARDS   T(I)                FORMAT E20,3
C CARD (N+1)     K1,K2,K3            FORMAT 3I10
C NEXT N CARDS(ONLY IF K2=3) A0(I)    FORMAT E20,3
C CODE NUMBERS MEANINGS
C   K1=1  THE PARENT ACTIVITY IS KEPT CONSTANT.
C   K1=2  THE PARENT ACTIVITY IS DECAYING.
C   K2=1  ONLY THE PARENT IS PRESENT AT T=0 WITH A UNIT ACTIVITY.
C   K2=2  ALL NUCLIDES ARE PRESENT AT T=0 WITH AN EQUAL UNIT ACTIVITY.
C   K2=3  THE INITIAL ACTIVITY OF EACH NUCLIDE IS SPECIFIED AS DATA.
C   K3=1  THE OUTPUT CONSISTS OF THE ABSOLUTE ACTIVITY OF EACH NUCLIDE.
C   K3=2  THE OUTPUT CONSISTS OF THE ACTIVITIES OF EACH NUCLIDE EXPRESSED
C          AS A FRACTION OF THE PARENT ACTIVITY AT THE GIVEN TIME
C   READ(5,1)N,M,TMIN,TMAX
C   1 FORMAT(2I10,2F10,3)
C   READ(5,2)(T(I),I=1,N)
C   2 FORMAT(E20,3)
C   READ(5,3)K1,K2,K3
C   3 FORMAT(3I10)
C   WRITE(6,15)N,M,TMIN,TMAX
C   15 FORMAT('0N=',I3,'X','M=',I2,'X','TMIN=',F10,3,'X','TMAX=',E10,3)
C   WRITE(6,16)K1,K2,K3
C   16 FORMAT('0CODE NUMBERS=',I10,3X,I10,3X,I10)
C   DO 100 I=1,N
C   100 LAM(I)=.693/T(I)
C   GO TO (94,95)K1
C   94 LAM(I)=0.
C   95 WRITE(6,11)
C   11 FORMAT('0 DECAY CONSTANTS,')
C   WRITE(6,12)(LAM(I),I=1,N)
C   12 FORMAT('0 *.10(F2),14/' *)
C   GO TO (96,97,98)K2
C   96 A0(I)=1.
C   DO 101 I=2,N
C   101 A0(I)=0.
C   GO TO 99
C   97 DO 102 I=1,N
C   102 A0(I)=1.
C   GO TO 99
C   98 READ(5,19)(A0(I),I=1,N)
C   19 FORMAT(E20,3)
C   99 WRITE(6,13)
C   13 FORMAT('0 INITIAL ACTIVITIES,')
C   WRITE(6,14)(A0(I),I=1,N)
C   14 FORMAT('0 *.10(E20,3/' *)
C   WRITE(6,4)
C   4 FORMAT('0 COEFFICIENTS OF THE DECAY SERIES,')

```

```

      B(1,1)=A0(1)
      WRITE(6,4)B(1,1)
9     FORMAT('0',E15,8)
      DO 300 I=2,N
      II=I-1
      DO 200 J=1,11
200    B(1,J)=LAM(1)/(LAM(1)-LAM(J))*H(11,J)
      SUM=A0(1)
      DO 201 J=1,11
201    SUM=SUM+B(1,J)
      B(1,1)=SUM
300    WRITE(6,5)(H(I,J),J=1,11)
      5     FORMAT('0',6(E15,8,3X)/' *',4(E15,8,3X))
      M1=M-1
      DT=(TMAX-TMIN)/M1
800    DO 600 L=1,M
      TIME=TMIN+(L-1)*DT
      GO TO(103,104)K1
103    DO 401 K=1,M
401    C(K)=EXP(-LAM(K)*TIME)
      GO TO 402
104    DO 400 K=1,N
400    C(K)=EXP((LAM(1)-LAM(K))*TIME)/A0(1)
402    A(1)=EXP(-LAM(1)*TIME)*A0(1)
      A(2)=C(1)*B(2,1)+C(2)*B(2,2)
      DO 500 I=3,N
      A(I)=C(1)*B(I,1)
      DO 500 J=2,11
500    A(I)=A(I)+C(J)*B(I,J)
      GO TO(501,502)K1
501    GO TO (701,702)K3
701    WRITE(6,17)
      17    FORMAT('0ABSOLUTE ACTIVITIES OF DAUGHTERS.0')
      GO TO 503
702    WRITE(6,6)
      6     FORMAT('0ACTIVITIES OF DAUGHTERS AS FRACTIONS OF PARENT.0')
      GO TO 503
502    GO TO (801,802)K3
801    WRITE(6,18)
      18    FORMAT('0FINAL ACTIVITY OF PARENT,ABSOLUTE ACTIVITY OF DAUGHTERS.0')
      GO TO 503
802    WRITE(6,10)
      10    FORMAT('0FINAL ACTIVITY OF PARENT,ACTIVITY OF DAUGHTERS AS FRACTIONS OF FINAL PARENT ACTIVITY.0')
503    WRITE(6,7)(A(I),I=1,M)
      7     FORMAT(' *',10(E21,10/' *'))
600    CONTINUE
      END

```

APPENDIX 4.1 - CHEMISTRY AND PHYSICAL STRUCTURE OF CONCRETE

Introduction

Before experimenting on the diffusion of Radon gas through concrete, it appeared necessary to gain some insight about the fine structure of what must be considered as a porous medium. This study should provide an idea of the orders of magnitude of the various parameters which are involved. Moreover, it will assist in familiarizing with the pore forming mechanisms that are involved, and should assist in conceiving adequate experiments to study Radon diffusion. We felt the necessity to rewrite the information obtained in three classic references [4.1, 4.7, 4.2] about concrete, because the intended use of the information is not the one of the authors. While the authors' primary concern was about the strength of the material and the permeability it would show toward liquids, our constant preoccupation in reading them was to get data about the physical structure of the material and its degree of permeability to a noble gas like Radon. The reader should keep in mind this total discrepancy in perspectives when sentences will be quoted from the sources used in writing this review.

This preliminary study shows the variability of the porosity of concrete as a function of

1. The water/cement ratio used in the mixing (w/c)
2. The handling of the mixture
3. The conditions of curing
4. The time allotted to curing under favorable conditions.

I. Concrete

Concrete can be considered to be made of two principal components, aggregate and paste. Aggregates are generally classified into two groups, fine and coarse. Fine aggregates consists of natural or manufactured and with particle sizes smaller than about 1/4 in.; coarse aggregates are those with particle sizes greater than about 1/4 in. The paste is composed in the case studied of Portland cement and water (sometimes entrained air). Figure 4.2 below shows volume proportions for two typical compositions of non-air entrained concrete.

Figure 4.2. From [4.1].

In properly made paste, each particle of aggregate is completely coated with paste. Also, all of the space between the aggregate particles is completely filled with paste (except for special concretes like air entrained concrete).

II. The Cement Paste

A. The Hydration Process

The report considers only Portland cement, the one most commonly used in the concrete mixtures for residential construction.

It is called a hydraulic cement for two reasons:

1. It needs only water for hardening to occur
2. The compound resulting from the reaction is water resistant.

Portland cement is an artificial mixture of mineral oxides. The mixture is finely ground to a powder called "dry cement," each grain of which consists of a prescribed mixture of the mineral oxides. When mixed with water, the dry cement turns into a paste with granular structure, made of the cement grains in suspension in the water. The configuration of the water can be visualized as a capillary system interconnecting all the water-filled spaces between the cement grains.

At the surface of each grain, some of the oxides react with the capillary water to form hydrated compounds which adopt a "gel-like" structure. This reaction of the oxides with water to form a gel is called the hydration of the cement. A "gel" is defined as: "an elastic, coherent mass consisting of a liquid in which ultramicroscopic particles are arranged in a fine network extending throughout the mass." Gels have very high specific surfaces (i.e., total surface of all the particles in a gram of the substance). Interesting electron microscope

pictures showing the progression of the formation of the gel structure in the capillary spaces are shown in Ref. [4.7], pages 32 through 36.

A subsequent hardening of this gel structure for reasons not presented in this elementary review provides the familiar hardened cement paste.

In order to derive quantitative predictions about the division of the total volume of the hardened paste among air, liquid, and solids it is necessary to study the hydration process and present some experimental data associated with the chemical reaction of hydration. These results will enable one to predict the volume breakdown in the solid cement.

B. Experimental Results About the Reactions Involved in the Hydration Process

The principal mineral oxides are the tricalcium silicate $(\text{CaO})_3 \cdot \text{SiO}_2$, the dicalcium silicate $(\text{CaO})_2 \cdot \text{SiO}_2$, the tricalcium aluminate $(\text{CaO})_3 \cdot \text{Al}_2\text{O}_3$, and the tetracalcium aluminoferrite $(\text{CaO})_4 \cdot \text{Al}_2\text{O}_3 \cdot \text{Fe}_2\text{O}_3$.

Their hydration is a multi-step reaction involving their hydrolysis. By convention all reactions are summarized under the term "hydration." Specialists in cement chemistry use the following short symbols for the oxides, a nomenclature that will be followed in this report.

$(\text{CaO}) = \text{C}$

$\text{SiO}_2 = \text{S}$

$\text{Al}_2\text{O}_3 = \text{A}$

$\text{Fe}_2\text{O}_3 = \text{F}$

Proportions of these four oxides in a typical Portland cement mixture are shown in Fig. 4.3 below (from Ref. [4.2], page 9).

<u>oxide</u>	<u>Cement</u>		
	1	2	3
CaO	66.	63.	66.
SiO ₂	20.	22.	20.
Al ₂ O ₃	7.	7.7	5.5
compound			
C ₃ S	65	33	73
C ₂ S	8	38	2
	73	71	75
C ₃ A	14	15	7
C ₄ AF	9	10	14

Figure 4.3 Percent by Weight of the Different Oxides in Typical Portland Cements

Notice the major role of the compounds C₃S and C₂S. They are responsible for the gel type structure. Since this report's interest is in the structure, the other oxides which are of less importance quantitatively will not be considered, nor shall the additives, the role of which is, for example, to control the hydration kinetics of the various compounds. The important factors to consider about the hydration products are:

1. The structure they adopt
2. The amount of capillary water involved in the hydration reactions
3. The change in the specific volume of the capillary water bound chemically in the hydrated products.

1. It is known that the hydrated crystals interlock in a "gel-like" structure. This structure evidently has pores which are much smaller than the capillary pores previously mentioned. These

are called "gel pores." It is essential to remember the difference between the capillary and gel pores. At the end of this section a summary will be made about what is known of each type.

2. The water in the gel structure is bound by different processes.

a) Some of the water is chemically bound to the solid grains of the gel under the form of hydrated products. The approximate hydration reactions of the principal oxides are the following, where the bracketed numbers indicate the amount of water chemically bound by the oxides as a percentage by weight of the respective dry oxides.

Notice that the tricalcium and dicalcium silicates result in the same hydrate.

b) Some of the water is absorbed at the surface of the hydrated compounds because of the large specific area of the gel and its colloidal behavior.

c) Finally, a portion of the water is trapped in the gel-pores. This water is differentiated from the capillary water for the very important reason that it cannot contribute any more to the hydration of the unhydrated oxides. The only site where hydration can take place is in the capillary pores and even there, the hydration actually stops whenever the capillary water has been used to the point where its vapor pressure drops to 0.8 of the atmospheric vapor pressure.

A method to compute the quantity of water that is bound chemically could be to use the percentage indicated with the hydration reaction and weight the terms in the summation by the proportion factors shown in Fig. 4.3. But the exact hydration reactions are not known and therefore another method is used: gel structures are submitted to drying conditions after full hydration of a cement paste. The amount of nonevaporable water (w_n) is estimated to be the chemically bound water, but it includes a certain amount of adsorbed water, the quantity depending on how drastically the drying is performed. These experiments performed under standardized conditions lead to the important result that the nonevaporable water typically represents 23 percent by weight of the dry cement.

Another important example of data that is obtained by this technique is the porosity of the gel. (For example, volume ratio between gel pore water and the total volume of the gel.) It is typically equal to 28 percent.

3. Finally, it has been found that the water reduces its specific volume by 0.254 during the hydration process. This reduction factor and the 28 percent porosity are fairly constant for any Portland cement.

The specific weight of dry cement is 3.15 g/cm^3 .

C. Assumptions Made in Predicting the Volume Partition of the Cement

Besides the experimental data mentioned, there are a few principles that one should consider when computing volume partitions.

1. Two types of laboratory curing are considered.

a) "Moist curing" during which water can always be supplied to the system through the capillary pores.

b) "Dry curing" which is assumed to take place in a sealed container thus allowing no outside interaction under the form of supply of water or evaporation.

2. The total volume available for the cured paste is equal to the total of the initial volumes of water and dry cement mixed together. This corresponds to a situation where there is no "bleeding" nor evaporation. This rule is as valid for dry as for moist curing whatever the extent of water addition in the latter process.

3. For the sake of simplicity, we will always assume in the dry curing process that hydration of the dry cement proceeds until all the capillary water is consumed even though we mentioned earlier that it actually stops when the vapor pressure of the water in the capillaries drops sufficiently.

D. Critical Values of Water/Cement (w/c)

We will consistently use the following symbols:

W = weight

V = volume

w = water

c = cement

gw = gel water

hw = hydration water, i.e., water to be used in the hydration but considered before the reaction has taken place

dc = dry cement

hc = hydrated cement

hs = hydrated substance, i.e., the sum of hydrated cement + gel water

w/c = water/cement ratio by weight

The two critical values we now want to establish are

1. The w/c ratio required to achieve full hydration of the dry cement involved in a mixture. In this situation, no oxide is to be found after hydration has taken place which has not combined with water to form one of the hydrates.

2. The w/c ratio required to achieve the same full hydration and obtain, besides, a hardened paste without capillary porosity. One will see that this latter requirement necessitates a moist curing process.

a) Water requirement for full hydration to occur. The amount of water (W_w or V_w) required to hydrate totally a given amount of dry cement (W_{dc} or V_{dc}) is independent of the hydration conditions; i.e., moist or dry curing. This necessary amount of water will include the water needed to perform the hydration reactions and the water contained in the gel pores which is not allowed to participate in the hydration reactions, since it is trapped in the gel pores of the hydrated substance. The computation is based on the experimental data given in paragraph B of this section.

i) It is known that the hydration water (W_{hw}) required to fully hydrate a given amount of dry cement (W_{dc}) amounts to 23 percent of the dry cement's weight. Using the metric system of units in which water has a specific weight equal to 1, it follows that

$$V_{hw} = W_{hw} = (.23)W_{dc} \quad (4.12)$$

ii) The specific weight of dry cement being 3.15 g/cm^3 , we have

$$W_{dc} = (3.15)V_{dc} \quad (4.13)$$

and therefore, by (1) and (2)

$$V_{hw} = (.23)(3.15)V_{dc} = (.725)V_{dc} \quad (4.14)$$

iii) The resulting volume of hydrated cement (V_{hc}) will be the original volume of the dry cement + the volume occupied by the water bound chemically. Recalling that water undergoes a reduction of specific volume (of 0.254) upon hydrating the cement,

$$V_{hc} = V_{dc} + (.746)V_{hw} \quad (4.15)$$

Substituting V_{hw} from (3) into (4) we get

$$V_{hc} = V_{dc} + (.746)(.725)V_{dc} = 1.54 V_{dc} \quad (4.16)$$

iv) The gel water occupies all the empty spaces in the gel structure and the porosity of the gel structure is equal to 28 percent. One can thus compute the volume of gel water (V_{gw}) given the volume of the hydrated cement (V_{hc}),

$$\begin{aligned} V_{gw} &= (.28)(V_{gw} + V_{hc}) \\ V_{gw} &= \frac{.28}{(1 - .28)} V_{hc} = .39 V_{hc} \end{aligned} \quad (4.17)$$

v) Substituting V_{hc} from (5) into (6), we obtain the volume of gel water trapped in the gel structure which resulted from the hydration of an initial volume of dry cement, V_{dc} .

$$V_{gw} = (.39)(1.54)V_{dc} = (.6)V_{dc} \quad (4.18)$$

The total volume requirement of water is therefore, by using (4.14) and (4.18),

$$V_w = V_{hw} + V_{gw} = (.725) + (.6)V_{dc} = 1.32 V_{dc} \quad (4.19)$$

vi) In terms of weight, (4.19) becomes

$$W_w = V_w = \frac{(1.32)W_{dc}}{3.15} = .42 W_{dc} \quad (4.20)$$

Equation (4.20) gives the first critical ratio we were looking for:

$$w/c = .42 \quad (4.21)$$

It is apparent that taking into account that part of the water which will not participate in the hydration process because it is trapped in the pores of the gel structure, it is necessary to mix 42 g of water with 100 g of dry cement to obtain full hydration of the cement.

b) Water required to obtain full hydration and a structure without capillary pores. Now compute the volume of capillary pores left empty of water in the case of a critical mixture of the first type, i.e., $w/c = .42$.

Recalling that if no bleeding nor evaporation occurs, the final volume of the hardened paste is equal to the sum of the original volumes of water and dry cement, and also assuming in the computation of the preceding critical ratio that all the water which has not been trapped in the gel pores has been used in hydration reactions, then if the volume of the hydrated substance ($V_{hs} = V_{hc} + V_{gw}$) does not occupy the total volume available, the difference is present in the cement paste under the form of an air-filled capillary system.

Assume 100 g of dry cement and 42 g of water which is submitted to dry curing.

i) The total volume (V_t) occupied by the hardened paste is

$$V_t = 42 + \frac{100}{3.15} = 42 + 31.8 = 73.8 \text{ cm}^3 \quad (4.22)$$

ii) The volume of the hydrated cement is, according to (4.16)

$$V_{hc} = (1.54)V_{dc} = (1.54)(31.8) = 49 \text{ cm}^3 \quad (4.23)$$

iii) The volume of the gel water is, according to (4.18)

$$V_{gw} = (.6)V_{dc} = (.6)(31.8) = 19.1 \text{ cm}^3 \quad (4.24)$$

iv) The total volume of the hydrated substance is thus by (4.23) and (4.24)

$$V_{hs} = 49 + 19.1 = 68.1 \text{ cm}^3 \quad (4.25)$$

But, as we have already pointed out, the hardened cement occupies 73.8 cm^3 (4.22). The difference is occupied by empty capillary pores which are of prime interest since they could be the preferred route for Radon diffusion.

The volume of the capillary pores amounts to

$$\text{Vol. of capillary pores} = (73.8 - 68.1) = 5.7 \text{ cm}^3$$

which represents

$$\frac{5.7}{73.8} \times 100 = 7.7\% \text{ of the volume of the hardened cement paste.}$$

As in the case of dry curing where full hydration is required, it follows that the initial volume of water and cement will not be completely filled by hydrated cement. In other words, the expansion of the cement upon hydration does not compensate exactly for the contraction of the water due to chemical combination.

This led to the problem of computing the initial volume that would lead to a hardened cement paste without capillary pores. Starting with the same amount of dry cement, it is necessary to add less water than

42 percent of its weight to decrease the total final volume, but if full hydration is required the missing amount of water must be provided during the curing process through the capillary pores. This is accomplished by performing the curing under moist conditions, i.e., under water.

To find the critical ratio w/c which amounts to exactly the final volume of completely hydrated cement, first note the relationships describing full hydration (see Eqs. (4.16) and (4.18)).

$$V_{hc} = (1.54)V_{dc} \quad (4.16)$$

$$V_{gw} = (0.6)V_{dc} \quad (4.18)$$

Next impose the condition that the final volume of hydrated substances $(V_{hc} + V_{gw})$ be equal to the initial volume of water and dry cement. Thus,

$$V_{dc} + V_w = V_{hc} + V_{gw} \quad (4.26)$$

Substituting V_{hc} and V_{gw} in (4.26) from (4.16) and (4.18),

$$V_{dc} + V_w = (.6)V_{dc} + (1.54)V_{dc}$$

or

$$V_w = 1.14 V_{dc} \quad (4.27)$$

In terms of weights,

$$W_w = \frac{1.14 W_{dc}}{3.15} = .36 W_{dc} \quad (4.28)$$

This second critical ratio ($w/c = .36$) leads to zero capillary porosity under moist curing conditions.

E. Pastes Resulting from Noncritical Values of w/c

Next consider excursions from the critical values in both directions for each of the curing processes and draw the resulting conclusions of interest to the diffusion problem.

1. w/c less than the critical value.

a) Dry curing. (critical value w/c = .42)

If $w/c < .42$ there will not be full hydration of the cement because of lack of water. Besides, there are still empty capillaries as the following computation shows.

Choose a mixture of $w/c = .3$

Starting with 100 g of dry cement and 30 g of water, the total volume available for the hardened cement paste is

$$30 + \frac{100}{3.15} = 30 + 31.8 = 61.8 \text{ cm}^3$$

The computation consists in finding how much dry cement will be hydrated. It is known from the preceding calculations (e.g., (12)) that 100 g of dry cement occupy 49 cm^3 after hydration. Call x the number of grams which will be hydrated in this case. They will occupy $(.49) x \text{ cm}^3$ after hydration. This $(.49) x \text{ cm}^3$ of hydrated cement represents the volume of the initial x grams of dry cement + the volume of the contracted water which has combined in the hydration of the x grams. Knowing that $(.23) x \text{ g}$ or cm^3 of water is required to hydrate x grams of dry cement, and also that those $(.23) x \text{ cm}^3$ of water undergo a volume contraction of $(.254)$ by combining with the oxides, the balance of the volumes must state that the initial volume of the x grams of dry cement plus the volume of the hydration water minus the contraction of the water has to equal the final volume

of the hydrated substance, i.e.,

$$\frac{x}{3.15} + (.23)x - (.23)(.254)x = .49x \quad (4.29)$$

or $x = 71.5$ grams of dry cement which corresponds to $71.5/3.15 = 22.7 \text{ cm}^3$ of dry cement. The amount of unhydrated cement is thus

$$31.8 - 22.7 = 9.1 \text{ cm}^3 \text{ or } 9.1 \times 3.15 = 28.7 \text{ g} \quad (4.30)$$

To find the final volume of the hydrated substance (V_{hs}) resulting from those 71.5 g of dry cement, add the volume of the hydrated cement (V_{hc}) to the volume of the gel water it contains.

$$V_{hc} = 71.5 \times (.49) = 35 \text{ cm}^3 \quad (4.31)$$

The volume of gel water (V_{gw}) is obtained from the expression of the porosity of the gel structure (28%), i.e.,

$$.28 = \frac{V_{gw}}{V_{hc} + V_{gw}} = \frac{V_{gw}}{35 + V_{gw}}$$

or

$$V_{gw} = 13.5 \text{ cm}^3 \quad (4.32)$$

Therefore,

$$V_{hs} = V_{gw} + V_{hc} = 13.5 + 35 = 48.5 \text{ cm}^3$$

The volume of empty capillaries is given by the difference between the total initial volume of dry cement and water minus the volumes of the hydrated substance and the unhydrated cement:

$$(31.8 + 30) - (48.5 + 9.1) = 4.2 \text{ cm}^3 \quad (4.33)$$

b) Moist curing. (critical value $w/c = .36$)

To reduce the computations, take the same mixture as in the preceding example: 100 g dry cement and 30 g of water. $w/c = .3$,

subcritical value. One can visualize a hydration process which would take place in two steps: First the same amount of dry cement is hydrated as in the preceding example, but since the curing is performed under moist conditions, the volume of empty capillaries which was computed is filled with water. Then the water is used to hydrate an additional amount of dry cement. This extra amount of dry cement will be such that the resulting hydration products (i.e., hydrated cement + gel water) will occupy the capillary pores that were filled with water plus the volume it occupied itself as dry cement before being hydrated. Now compute the extra $y \text{ cm}^3$ of cement which will be hydrated to occupy the 4.2 cm^3 of capillary spaces given by Eq. (4.33) + the $y \text{ cm}^3$ they were occupying as dry cement. It is known that 100 g (31.8 cm^3) of dry cement gives rise to 68.1 cm^3 of hydrated substance (Eq. (14)) which corresponds to a change in volume of $\frac{68.1}{31.8} = 2.14$. Thus the extra $y \text{ cm}^3$ of cement are going to occupy $y(2.14) \text{ cm}^3$. The balance of volumes gives

$$\frac{y(2.14) + y}{y} = 2.14$$

or

$$y = 3.7 \text{ cm}^3 \quad (4.34)$$

The amount of still unhydrated cement is then

$$9.1 - 3.7 = 5.2 \text{ cm}^3$$

and there is no capillary porosity any more.

c) Conclusions

It is concluded from these two subcritical mixtures that they both lead to a certain degree of unhydrated cement and only the moist curing provides a hardened paste without capillary porosity.

2. w/c greater than critical values

For economical and convenience reasons, this case is the one which typically occurs during construction. Compute the partition of the volume that this condition leads to.

a) Dry curing. (critical value $w/c = .42$)

Take a case slightly supercritical, i.e., $w/c = .475$.

This is the case of a mixture of 126 g of dry cement ($W_{dc} = 126 \text{ g}$, $V_{dc} = 40 \text{ cm}^3$) with 60 g of water ($W_w = 60 \text{ g}$, $V_w = 60 \text{ cm}^3$). The total final volume of the paste will be

$$V_t = 40 + 60 = 100 \text{ cm}^3$$

Since complete hydration of the cement will certainly take place, by (4.16) we have

$$V_{hc} = (1.54)V_{dc} = (1.54) 40 = 61.6 \text{ cm}^3$$

The volume of the gel water is obtained by expressing the porosity of the gel structure

$$\frac{V_{gw}}{61.6 + V_{gw}} = .28$$

or

$$V_{gw} = 24 \text{ cm}^3$$

The total volume of the hydrated substance is thus

$$V_{hs} = 61.6 + 24 = 85.6 \text{ cm}^3$$

The amount of capillary volume is thus

$$V_{cap} = 100 - 85.6 = 14.4 \text{ cm}^3$$

which corresponds to 14.4 percent of the volume of the hardened cement.

Only part of those capillaries will be occupied by the water which was not used in the hydration. Now compute how much water was used. It is equal to the hydration water + the gel water

$$V_{hw} = V_{gw} = (.23)126 + 24 = 29 + 24 = 53 \text{ cm}^3$$

Therefore the volume of unused water is

$$60 - 53 = 7 \text{ cm}^3$$

This water is found in the 14.7 cm^3 of capillaries and we thus have

$$14.4 - 7 = 7.4 \text{ cm}^3 \text{ of empty capillaries.}$$

b) Moist curing. (critical value $w/c = .36$)

By choosing the same extracritical mixture as in the above example one will get the same volume partition between cement paste and capillaries, since full hydration had already occurred under dry curing. The difference resides in the fact that under moist curing all the capillaries are filled with water.

c) Conclusion

In the case of supercritical values of w/c there will result a significant porosity (14.4%) for an increase in water the content of $0.36 - 0.47 = 0.11$ in the absolute value above the critical value of the mixture which leads to zero capillary porosity. The actual increase of routes for the Radon will depend on the facility of the remaining capillary water we encountered in these last two examples to evaporate from the cement under atmospheric conditions.

3. General conclusion about noncritical mixtures

The amount of capillary porosity depends not only on the ratio w/c but also on the conditions under which curing has been

performed. Super-critical values of the mixture can lead to a zero capillary porosity only if the curing is moist.

In order to compare the theoretical curing conditions assumed in this study to the actual on site conditions one has to take into account the amount of water that evaporates from the freshly poured mixture before having a chance to enter the early setting process which determines the final volume of the hardened paste. Also, moist curing was supposed to be of the immersed type whereas construction practice consists of keeping the surfaces of the concrete moist by spraying water intermittently or by covering them with moisture-retaining material such as burlap. Finally, it will often occur that real curing will be a combination of moist curing during the first period of curing followed by a dry curing which no water is added but it evaporates from the capillaries.

F. The Pores and the Segmentation of the Capillary System

Two types of pores are to be considered:

1. The gel pores.

The structure of the gel can be described as a network of bundles of fibrous particles giving rise to a system of interconnected interstitial spaces filled with water.

The water contained in the gel pores is evaporable to various extents as has been discussed.

The diameters of the pores vary between $15-20 \text{ \AA}$ (10^{-8} cm) (i.e., approximately 10 times bigger than water molecules). They represent 28 percent of the total volume of the hydrated substance. As the hydration (curing) proceeds they fill the space occupied by the capillary pores.

2. Capillary pores

a) the pore itself.

Varies greatly in shape, but its dimensions estimated from water vapor pressure are of the order of 2×10^{-5} cm.

b) the capillary system.

During curing the capillary pores usually form an interconnected system randomly distributed throughout the concrete paste. As the curing progresses, there is a possibility of segmenting the capillary system by the gel so that the only connection remaining between capillary pores are gel pores. This segmentation is extremely important with respect to the porosity of the cement paste and is a function of

i) w/c

ii) degree of curing achieved.

The degree of curing achieved is itself a function of

i) total duration of curing

ii) type of cement used.

Figure 4.4 shows the stage at which the capillary system ceases to be continuous as a function of the w/c. This is valid for all Portland cement. Figure 4.5 shows the time of moist curing required. This is a function of the type of cement used.

Conclusion

1. Importance of w/c ratio on amount of capillary porosity.
2. Evolution of the porosity during curing by shifting from open capillary porosity to segmented capillary porosity and finally to complete gel porosity.

3. Importance of curing time under favorable conditions to obtain segmentation.

4. Importance of the fate of the capillary water in the post curing period in providing preferred route for the Radon diffusion.

Figure 4.4 From [4.2]. Relation between the Water/Cement Ratio and the Degree of Hydration at Which the Capillaries Cease to be Continuous.

APPROXIMATE TIME REQUIRED TO PRODUCE MATURITY AT WHICH CAPILLARIES BECOME SEGMENTED

Water/cement ratio by weight	Time required
0.40	3 days
0.45	7 days
0.50	14 days
0.60	6 months
0.70	1 year
over 0.70	impossible

Figure 4.5 from [4.2]. Approximate Time Required to Produce Maturity at Which Capillaries Become Segmented

Notice that for $w/c = 0.7$ segmentation will never be obtained (except in the special case of very fine cement).

III. Bleeding (From [4.7])

The term "bleeding" or "water separation" denotes the capacity of fresh concrete or mortar to undergo sedimentation during the period preceding setting. The solid particles in the concrete mix tend to settle and a part of the mixing water is displaced and collects at the surface of the freshly placed concrete. Naturally, this settling of fresh concrete tends to decrease the solid volume of the hardened concrete. Thus, the "bleeding" of concrete might be regarded as desirable, because it must lead to some reduction in the w/c ratio. There are, however, other deterring factors, since bleeding concurrently disturbs the internal homogeneity of concrete. The mixing water, rising to the surface, carries away the finest particles of cement with the result that, in extreme cases, a water-rich cement slurry of corresponding low strength collects at the surface of the concrete, while at the same time flow channels with low cement content are formed in the body of the concrete. Furthermore, this segregation of the mixing water is not limited to the surface of the concrete only; the coarse aggregate particles form a rigid skeleton of mutually supporting units so that their settling stops. However, sedimentation goes on within this framework. The result is shown schematically in an exaggerated sketch in Fig. 4.6. The fine-grained mortar has settled from underneath the larger aggregate particles and its place is now occupied by a water-rich slurry of low strength. Similar phenomena can occur in reinforced concrete where the settling movement

causes a space beneath the reinforcement. The bond to the reinforcement is then weakened. This is undesirable, not only for bond strength, but also because of the danger of corrosion of the reinforcement at the void.

Schematic representation of "bleeding."

Figure 4.6. (From [4.7])

Bleeding is most likely to occur in concretes that are poor in fine sand or when too coarse a cement has been used. This can also occur when the concrete mix is too lean or the use of too much mixing water. In addition, bleeding is caused, to some extent, by some specific properties of cement, e.g., delayed initial settling will prolong the time in which sedimentation can occur until the setting of the paste terminates this form of segregation. Care, however, must be taken in applying measures to prevent concrete bleeding. The use of too much finely ground cement is not always desirable, nor is too short a period of setting time. An addition to concrete materials

rich in fines can often do more harm than good because the stabilizing effect thus produced is achieved at the expense of workability or a sufficiently low w/c ratio. As is often the case in concrete technology, a compromise solution must be found. Sometimes the use of correct pouring technique can eliminate the adverse effects of concrete bleeding; if at the end of bleeding and at the beginning of setting, the freshly poured concrete is subjected to vibration or tamping, the homogeneity of the mix is restored and the quality of the finished concrete shows a surprising improvement.

IV. The Aggregates

A look at the porosity and permeability to water.

A. Porosity

The coarse aggregates represent from 30 to 50 percent of the volume of the concrete. Their contribution is an important one to the overall porosity of the concrete.

The smallest pores of the aggregate are larger than the gel pores in the cement gel. Some of the pores open at the surface of the aggregate, some others are wholly inside (no information available about interconnection).

<u>ROCK</u>	<u>POROSITY (%)</u>
Gritstone	0 - 48.
Quartzite	1.9 - 15.
Limestone	0 - 37.
Granite	.4 - 3.8

Fig. 4.7 Order of Magnitude of Porosity of Common Rocks.

B. Permeability to Water

It is interesting to report comparisons which have been made between the permeability to water (for definition see Part V) of cement paste compared to some aggregates.

COMPARISON BETWEEN PERMEABILITIES OF ROCKS AND CEMENT PASTES

Type of rock	Coefficient of permeability cm/s	Water/cement ratio of mature paste of the same permeability
Dense trap . .	2.47×10^{-12}	0.38
Quartz diorite	8.24×10^{-12}	0.42
Marble	2.39×10^{-11}	0.48
Marble	5.77×10^{-10}	0.66
Granite	5.35×10^{-9}	0.70
Sandstone . . .	1.23×10^{-8}	0.71
Granite	1.56×10^{-8}	0.71

Fig. 4.8 from [4.2]. Comparison Between Permeabilities of Rocks and Cement Pastes.

V. Permeability of Concrete to Water

As mentioned before, this part is reported here as a typical study of permeability of concrete. The major differences between this study and the one being considered is:

1. Water is a liquid while Radon is a gas which results in a large difference in the order of magnitude of the mean free paths of the diffusing substances.

2. In the early stages of curing, water is a component of the concrete and part of the water constituting the concrete participates in the flow of the water through the concrete. Radon amounts to very minute quantities in the aggregate.

Definition of Permeability Coefficient "K"

It measures the quantity of water flowing through a given thickness under a given pressure. K is defined by Darcy's equation

$$\frac{dg}{dt} = \Delta K \frac{\Delta h}{L}$$

where

$$\frac{dg}{dt} = \text{rate of flow } (\text{cm}^3/\text{s})$$

$$A = \text{surface area of specimen } (\text{cm}^2)$$

$$K = \text{permeability coefficient } (\text{cm/s})$$

$$h = \text{drop in hydraulic head } (\text{cm})$$

$$L = \text{thickness of sample } (\text{cm})$$

Notice that Darcy's law is exactly the application of Fick's law of diffusion relating the flux to the driving force. It appears thus that the continuum concept was applied to the diffusion of water through concrete.

Determining Factors in the Permeability to Water

Both cement paste and aggregate contain pores. Besides those, other voids may appear due to incomplete compaction and bleeding. These voids range from a fraction of a percent to 10 percent, the last figure corresponding to bad concrete.

Nevertheless, in fully compacted concrete, it is the permeability of the cement paste that has the greatest effect on the overall permeability of the concrete.

The porosity of the paste can be described by the

gel pores: 28 percent of gel paste .

capillary pores: 0 - 40 percent depending on the w/c and degree of hydration.

Experiments show that it is the capillary porosity which controls the permeability to water. Values reported for K are 7×10^{-14} cm/s for cement gel and 20 to 100 times more for the cement paste as a whole including the capillary pores.

Experiments have been performed to show the influence of different factors on the permeability of water, principally curing, capillary porosity, w/c, fineness of cement grains and the compound composition.

1. Curing

Fresh paste: the flow is controlled by size and shape of original grains of cement.

During hydration: permeability decreases as gel takes the place of the space filled by capillary water.

Mature paste: the permeability depends now on

- a. size, shape, concentrations of gel particles
- b. whether or not capillary pores have been segmented by cement gel.

REDUCTION IN PERMEABILITY OF CEMENT PASTE (WATER/CEMENT RATIO = 0.7) WITH THE PROGRESS OF HYDRATION

Age days	Coefficient of permeability, K cm/s
Fresh	2×10^{-4}
5	4×10^{-8}
6	1×10^{-8}
8	4×10^{-9}
13	5×10^{-10}
24	1×10^{-10}
ultimate	6×10^{-11} (calculated)

Fig. 4.9 from [4.2]. Reduction in Permeability of Cement Paste (Water/Cement Ratio = 0.7) With the Progress of Hydration.

2. Capillary Porosity

See Fig. 4.10. Notice that K is multiplied by 100 for porosity going from 10 percent to 38 percent.

Figure 4.10. (From [4.2]) Relations Between Permeability and Porosity of Cement Paste.

Figure 4.11. (From [4.2]). Relation Between Permeability and Water/Cement Ratio for Mature Cement Pastes (93 Per Cent of Cement Hydrated).

Notice the steep increase in the line for w/c 0.6, when the capillary pores are not segmented any more.

4. Fineness of Cement

Coarse cement results in more porous cement.

5. Compound Composition

No effect ultimately but affects kinetics and thus values of permeability as a function of time.

APPENDIX 5.1 - ORDER-OF-MAGNITUDE ANALYSES FOR THE RECORDING OF LOCAL RADON FLUXES

1. Necessity for a Method to Record Local Fluxes

The solution to problems created by the effusion of Radon into enclosed areas often requires estimates of the Radon fluxes through specific locations or during specified periods of time. The proposed method of local flux determination consists in the positioning of a sealed container over a fraction of the area to be monitored. The initial Radon concentration in the container and the concentration built over a time " T_s " are then recorded and processed to evaluate the Radon flow through the particular area during the time period " T_s ."

2. Need for a Theoretical Analysis

Sources of error are theoretically introduced by the sampling method described above because of the modifications introduced in the Radon flow pattern in the vicinity of the monitored area by the presence of the Radon concentration buildup in the sampling container. Therefore, an analysis is required to derive design criteria for the experimental set-up in order to reduce to a reasonable fraction the approximations involved by using a linear formulation of the following type:

$$\phi = \frac{(C_f - C_i)}{T_s} \frac{V}{A} \quad (5.60)$$

where

ϕ = Flow per unit area and unit time

C_i = Initial concentration in sampling container

C_f = Final concentration in sampling container

T_s = Time lag between C_i and C_f

V = Volume of sampling container

A = Area covered by sampling container

As will be shown, the use of a formulation as simple as (5.60) imposes restrictions on the extent of the sampling time T_s . It also requires that the lateral dimensions of the sampling device be large enough to minimize the error introduced by the flow bypassing the edges of the container. Other restrictions are imposed on the maximum volume of the container if a measurable concentration has to build up in the container over a period T_s . Finally, the volume of the sampling container should be large enough to limit the perturbations introduced when the atmospheres of the sampling container and of the Radon scintillation chamber (with its auxilliary tubing) are mixed for measurement purposes. As order-of-magnitude arguments are presented, it should be recalled that the purpose of the following analysis is not to obtain exact values of physical quantities, but rather to help in establishing design criteria for an experimental set-up.

3. Governing Equations

In absence of any conclusive evidence on possible major effect of atmospheric conditions on the Radon flow rate in dwellings, a linear model of diffusion is assumed to apply to the concentration-driven Radon exchanges. The total flux ϕ_T through an area A is then given by

$$\phi_T(t) = -k A [\text{grad } C(x,t)]_{x=0} \quad (5.61)$$

where

$x = 0$ specifies the location of the monitored area

k = effective diffusion coefficient of the permeated medium

$C(x,t)$ = Radon concentration in the medium.

By insertion of (5.61) into a balance equation for a differential volume of the medium, the complete equation describing the concentration driven diffusion of a radioactive gas like Rn-222 is

$$\nabla^2 C(x,t) - q^2 C(x,t) = \frac{1}{k^*} \frac{\partial C(x,t)}{\partial t} \quad (5.62)$$

where

$$q = \sqrt{\lambda/k^*}$$

λ = decay constant of the radioactive gas

k^* = k /porosity

According to the expected behavior of the flow pattern, only two spatial coordinates need be introduced. The first one (r or y) will extend in the direction of the lateral extent of the monitored area. The second coordinate will extend perpendicularly to the first one, in a direction parallel to the original Radon flow.

In order to facilitate order of magnitude analyses, Eq. (5.62) is made nondimensional by the introduction of the following new variables

$$C^+(x,t) = C(x,t)/C_M \quad (5.64)$$

$$\tau^+ = \lambda t \quad (5.65)$$

$$x_i^+ = x_i/l_i \quad (5.66)$$

where

C_M = maximum Radon concentration encountered in the experiment

x_i = any of the two spatial variables

l_i = characteristic length in the direction of the variable x_i

By introducing (5.64), (5.65), (5.66) into (5.62) we obtain, in cylindrical coordinates

$$\frac{1}{l_r^2} \frac{1}{r^+} \frac{\partial}{\partial r^+} \left(r^+ \frac{\partial C^+}{\partial r^+} \right) + \frac{1}{l_z^2} \frac{\partial^2 C^+}{\partial z^{+2}} - q^2 C^+ = \frac{\lambda}{k^*} \frac{\partial C^+}{\partial t^+} \quad (5.67)$$

and in Cartesian coordinates

$$\frac{1}{l_y^2} \frac{\partial^2 C^+}{\partial y^{+2}} + \frac{1}{l_z^2} \frac{\partial^2 C^+}{\partial z^{+2}} - q^2 C^+ = \frac{\lambda}{k^*} \frac{\partial C^+}{\partial t^+} \quad (5.68)$$

where

l_r, l_y, l_z = characteristic lengths.

The treatment of the cylindrical and the Cartesian cases are identical and we will henceforth refer to Eq. (5.68) only.

4. Order-of-Magnitude Analysis

Radon diffusion experiments performed on concrete walls yielded values of k^* of the order of $5 \times 10^{-5} \text{ cm}^2/\text{sec}$. In ordinary construction projects related to dwellings it is rare to encounter concrete thicknesses larger than about 20 cm. This limits the magnitude of l_z . If we multiply (5.68) by l_z^2 we obtain

$$\left(\frac{l_z}{l_y}\right)^2 \frac{\partial^2 C^+}{\partial y^{+2}} + \frac{\partial^2 C^+}{\partial z^{+2}} - l_z^2 q^2 C^+ = \frac{l_z^2 \lambda}{k^*} \frac{\partial C^+}{\partial t^+} \quad (5.69)$$

If the lateral extent (l_y) of the sampled area is about 3.3 times as large as l_z , the first term in (5.69) becomes of the order 10^{-1} , while the second term is of order 1. An error limited to 10 percent would then be made by assuming the problem to be one dimensional. The order of the third term which describes the importance of the decay in the diffusion process is determined by

$$\frac{l_z^2 \lambda}{k^*} = \frac{4 \times 10^2 \times 2.1 \times 10^{-6}}{5 \times 10^{-5}} = 16.8$$

and cannot be neglected.

The time dependent term is of the same order of magnitude as the decay term and also has to be conserved.

Since we have established the criteria to be adhered to in order to treat the flow into the sampling container as a one dimensional case, the only reason for studying the solution of the remaining equation, i.e.,

$$\frac{d^2 C^+}{dz^2} - (k_2 q)^2 \cdot C^+ = (k_2 q)^2 \frac{\partial C^+}{\partial t} \quad (5.70)$$

would be to establish the time dependence of the flux into the container as the concentration builds up in the latter. The time dependence of the flux would allow us to estimate the time " T_s " after which it would become unreasonable to consider the flux to be practically equal to the original flux. A simpler method of evaluating this limit " T_s " consists of computing the ratio of the steady state fluxes in the two extreme cases, i.e., at the beginning of the experiment and after a steady state situation has been reached which maintains a steady concentration in the container. The magnitude of the flux encountered during the time dependent experiment falls between these two extreme fluxes, and if the two steady state fluxes do not differ appreciably, the assumption of a constant average flux during the sampling time " T_s " should be a valid one.

The steady state case is governed by

$$\frac{d^2 C}{dz^2} - q^2 C = 0 \quad (5.71)$$

In the case depicted by Fig. 5.2 of a diffusion taking place between a high concentration C_s (concentration in the tailings) and a low

concentration C_c (concentration in container C_i or C_f) the solution of (5.71) is

$$C(x) = \frac{C_c \sinh q(T-x) + C_s \sinh (qx)}{\sinh (qT)} \quad (5.72)$$

Application of (5.61) to (5.72) gives the flux into the container

$$\phi = k \left. \frac{dC}{dx} \right|_{x=0} = \frac{kq [C_s - C_c \cosh(qT)]}{\sinh (qT)} \quad (5.73)$$

If we compare the fluxes into the container in presence of the two concentrations C_i and C_f we obtain

$$\frac{\phi_i}{\phi_f} = \frac{C_s - C_i \cosh (qT)}{C_s - C_f \cosh (qT)} = \frac{1 - C_i/C_s \cosh (qT)}{1 - C_f/C_s \cosh (qT)}$$

with $T = 2 \times 10^1$ and $q = \sqrt{\frac{\lambda}{k^*}} = \sqrt{\frac{2.1 \times 10^{-6}}{5 \times 10^{-5}}} = .2$,

we obtain

$$qT = 4 \quad \text{and} \quad \cosh(qT) = 27.3 .$$

Concentrations observed in the field average around 40 pCi/cm^3 for C_s , 0.05 pCi/cm^3 for C_i and 0.3 pCi/cm^3 for C_f . These numbers lead to a ratio

$$\frac{\phi_i}{\phi_f} = \frac{1 - (27.3 \times 0.05/40)}{1 - (27.3 \times 0.3/40)} = \frac{.966}{.795} = 1.2$$

The reduction in flux would therefore reach 20 percent if the experiment was pursued until an equilibrium point was reached. The maximum value intended for " T_s " is the minimum time necessary between two consecutive measurements. This time amounts to approximately 4 hours. It is one order of magnitude less than the recorded times necessary for the

concentration to evolve from C_i to C_f . We will therefore assume a constant average flux in experiments relative to concrete surfaces.

5. Correction of the Readings

The concentrations C_i and C_f are obtained by circulating the container's atmosphere into a Radon scintillation chamber. The atmosphere of the scintillation chamber (135 ml + 3 ml tubing) contains a very low concentration in Radon (≈ 5 pCi/liter at most). The atmosphere existing in the manual rubber bulb pump and its associated filter and tubing (98 ml) contains a higher Radon background because it is flushed on the sampling location inside experimental buildings where the Radon level is high (≈ 70 to 150 pCi/liter). After these three atmospheres have been mixed for the purpose of measuring C_i or C_f , the measured concentration (C_m) is not the actual concentration (C_a) which existed in the sampled container, but a diluted one. The correction is performed in the following manner. First an average background concentration C_b is calculated. It results from the mixture of the scintillation chamber volume with the pump and is given by

$$C_b = \frac{5 \times 138 + 98 \times C'_b}{236}$$

where C'_b = concentration recorded at the experimental site at the time of measurement.

Next we consider that the total number of Radon atoms (n_a) in the container (volume = V_c) before the measure amounts to a concentration

$$C_a = \frac{n_a}{V_c}$$

Similarly the total number of Radon atoms (n_b) in the measuring device (Radon chamber + pump = V_{aux}) generate a concentration

$$C_b = \frac{n_b}{V_{aux}}$$

The measured concentration C_m results from the mixture of $n_a + n_b$ in a volume $V_c + V_{aux}$, i.e.,

$$C_m = \frac{n_a + n_b}{V_c + V_{aux}} \quad (5.76)$$

Dividing numerator and denominator by V_c we have

$$C_m = \frac{C_a + n_b/V_c}{1 + V_{aux}/V_c} \quad (5.77)$$

Rearranging (5.77) we get

$$C_a = C_m + (C_m - C_b) \frac{V_{aux}}{V_c} \quad (5.78)$$

The time laps " T_s " extends from $t = 0$ to $t = t_1$.

If, besides C_i and C_f , a third measurement is made after a second lapse of time, T_s^1 extending from $t = t_1$ to $t = t_2$, the reference concentration to be used in evaluating the flux during this extended period should be the measured concentration at $t = t_1$ and not the actual concentration at that time.

Finally, because of the dilution introduced by the measurement at $t = t_1$, the average flux between the time $t = 0$ and the third measurement ($t = t_2$) should be obtained by a weighted average between the fluxes recorded during the two consecutive accumulation periods, rather than by a comparison of the concentrations provided by the initial and third measurements.

Appendix 5.2. Program KCANFLU. Solution of Transcendental Equation by Newton's Algorithm.

```

PROGRAM KCANFLU(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)
DIMENSION Y(100)
SINH(X)=(EXP(X)-EXP(-X))/2.
COSH(X)=(EXP(X)+EXP(-X))/2.
READ(5,1) T,CS,CC,F,PORC
WRITE(6,9) T,CS,CC,F,PORC
9 FORMAT('0T=*,E10.3,3X,*CS=*,E10.3,3X,*CC=*,E10.3,3X,*F=*,E10.3,3X,*
Z*PORC=*,E10.3)
F=F/PORC
1 FORMAT('E20.3)
RADOLAM=.493/(3.62*8.64E+04)
SORA=SQRT(RADOLAM)
A=SORA*CS
B=SORA*CC
C=T*SORA
Y(1)=2.E+03
WRITE(6,8) Y(1)
8 FORMAT('0INITIAL GUESS,Y(1)=*,E21.14)
DO 3 I=2,100
M=I
X=C/Y(I-1)
RAT=(A*Y(I-1)-R*Y(I-1)*COSH(X)+F*SINH(X))/(A+B*C*SINH(X)/Y(I-1)+
)COSH(X)*(F*C/Y(I-1)/Y(I-1)-H))
Y(I)=Y(I-1)-RAT
WRITE(6,2) Y(I)
2 FORMAT('0Y=*,F21.14)
CRIT=ABS((Y(I)-Y(I-1))/Y(I))
IF(CRIT.LE.1.E-05)GO TO 99
3 CONTINUE
WRITE(6,4)
4 FORMAT('0CRITERIA NEVER SATISFIED*)
GO TO 6
99 WRITE(6,5)
5 FORMAT('0CRITERIA SATISFIED*)
6 CONTINUE
AKSTAR=Y(M)*Y(M)
WRITE(6,10) AKSTAR
10 FORMAT('0AKSTAR=*,E20.13)
AK=AKSTAR*PORC
WRITE(6,7) AK
7 FORMAT('0DIFFUSION COEFFICIENT=*,E21.14)
END

```

Appendix S.3. Program BASEFLUO. Distribution of Radon Concentrations and Fluxes Across Multilayered Porous Medium. No Sealant Case.

```

PROGRAM BASEFLUO(INPUT,OUTPUT,TAPE4=INPUT,TAPE6=OUTPUT)
DIMENSIONZ(101),Z2(101),Z3(101),C1(101),C2(101),C3(101),F1(101),F
22(101),F3(101)
SINH(X)=(EXP(X)-EXP(-X))/2.
COSH(X)=(EXP(X)+EXP(-X))/2.
READ(5,*)HA,R0,F
1 FORMAT(3F20.3)
WRITE(6,2)RA,R0,F
2 FORMAT(*0HA=*,F10.3,*GW HA/GW TAILIGS*,3X,*R0=*,E10.3,*GR/CM3*,3X,
2*F=*,E10.3)
READ(5,3)AK1,AK2,AK3
3 FORMAT(3E20.3)
WRITE(6,4)AK1,AK2,AK3
4 FORMAT(*0AK1=*,E10.3,*CM2/SEC*,3X,*AK2=*,E10.3,*CM2/SEC*,3X,*AK3=*,
2,E10.3,*CM2/SEC*)
READ(5,30)PORS,PURT,PORC
30 FORMAT(3F20.3)
WRITE(6,33)PORS,PURT,PORC
33 FORMAT(*0PORS=*,E10.3,3X,*PURT=*,E10.3,3X,*PORC=*,E10.3)
READ(5,5)TH1,TH2
5 FORMAT(2E20.3)
READ(5,7)CA
7 FORMAT(F20.3)
WRITE(6,8)CA
8 FORMAT(*0ATMOSPHERIC CONCENTRATION=*,E20.3,*PCI/CM3*)
READ(5,9)N1,N2,N3
9 FORMAT(3I10)
S=RA*R0*F*2.1E-06/VORT
P1=SQRT(2.1E-06/AK1)*PORS
P2=SQRT(2.1E-06/AK2)*PURT
P3=SQRT(2.1E-06/AK3)*PORC
D1=1./P1
D2=1./P2
D3=1./P3
TH3=7.*D3
WRITE(6,20)TH1,TH2,TH3
20 FORMAT(*0TH1=*,F10.3,3X,*TH2=*,F10.3,3X,*TH3=*,F10.3)
WRITE(6,11)S,D1,D2,D3
11 FORMAT(*0SOURCE=*,E10.3,*PCI/CM3,SEC*,3X,*RELAXATION DISTANCES,
201=*,E10.3,*CM*,3X,*D2=*,E10.3,*CM*,3X,*D3=*,E10.3,*CM*)
WRITE(6,10)N1,N2,N3
10 FORMAT(*0*10,*STEPS IN REGION 1*,3X,*10,*STEPS IN REGION 2*,3X,*1
210,*STEPS IN REGION 3.*)
T1=TH1
T2=TH1+TH2
PT1=P1*T1
PT2=P2*T2
X1=SINH(PT1)
X2=-COSH(PT1)
X3=S/2.1E-06
X4=AK1*P1
X5=-AK1*P1*COSH(PT1)
X6=AK1*P1*SINH(PT1)
X7=AK1*P1
X8=-AK2*P2*COSH(P2*TH2)
X9=-CA*SINH(1/2*TH2)*AK2*P2
X10=SINH(P2*TH2)
X11=CA*COSH(P2*TH2)-X7
D=-X5*X8-X7*X4*X10+X4*(X1*X8+X7*X2*X10)
DA34=-X8*(X1*X5+X11*(X1*X6-X5*X2))-X10*(-X3*X6*X7+X9*(X1*X6-X5*X2))
21

```

```

DA14=-X9*X6*X10+X11*X4*XH-X4*(-XH*(X3-X2*X1))-X10*X9*X2)
DB14=X5*(X9*X10-X11*XH)-X4*(X1*(X9*X10-X11*XH)-X/*X3*X10)
DA24=-X5*X9-X7*X6*X11-X4*(-X1*X9-X7*(X2*X11-X3))
A34=DA34/D
A14=DA14/D
B14=DB14/D
A24=DA24/D
WRITE (A,32) A34,A14,B14,A24
32 FORMAT ('0A34=*,E10.3,*A14=*,E10.3,*B14=*,E10.3,*A24=*,E10.3)
WRITE (A,21)
21 FORMAT ('0FLUXES')
F13=AK1*P1*(A14*COSH(P1))-B14*SINH(P1))
F12=AK1*P1*A14
F4=-AK2*P2*A24
WRITE (B,22) F13,F12,F4
22 FORMAT ('0F13=*,E10.3,3X,*F12=*,E10.3,3X,*F4=*,E10.3)
WRITE (B,28)
28 FORMAT ('0CONCENTRATION DISTRIBUTIONS.')
DX1=TH1/(N1-1)
DX2=TH2/(N2-1)
DX3=TH3/(N3-1)
WRITE (A,23) DX1,DX2,DX3
23 FORMAT ('0STEP SIZES,DX1=*,F10.3,*CM*,3X,*DX2=*,F10.3,*CM*,3X,*DX3=
*,F10.3,*CM*)
31 FORMAT ('0*,5X,*LOCATION*,17X,*CONCENTRATION*,17X,*FLUX*)
WRITE (A,24)
24 FORMAT ('0CONCENTRATION DISTRIBUTION IN THE TAILINGS.')
WRITE (A,31)
Z1(1)=0.8
C1(1)=A14*SINH(P1*(Z1(1)-T1))+B14*COSH(P1*(Z1(1)-T1))+S/2.1E-06
F1(1)=-AK1*P1*(A14*COSH(P1*(Z1(1)-TH1))+B14*SINH(P1*(Z1(1)-TH1)))
WRITE (A,25) Z1(1),C1(1),F1(1)
25 FORMAT ('E15.5,10X,E15.5,10X,F15.5)
DO 100 I=2,N1
Z1(I)=Z1(I-1)+DX1
C1(I)=A14*SINH(P1*(Z1(I)-T1))+B14*COSH(P1*(Z1(I)-T1))+S/2.1E-06
F1(I)=-AK1*P1*(A14*COSH(P1*(Z1(I)-TH1))+B14*SINH(P1*(Z1(I)-TH1)))
100 WRITE (A,25) Z1(I),C1(I),F1(I)
WRITE (A,26)
26 FORMAT ('0CONCENTRATION DISTRIBUTION IN CONCRETE.')
WRITE (A,31)
Z2(1)=T1
C2(1)=A24*SINH(P2*(Z2(1)-T2))+CA*COSH(P2*(Z2(1)-T2))
F2(1)=-AK2*P2*(A24*COSH(P2*(Z2(1)-T2))+CA*SINH(P2*(Z2(1)-T2)))
WRITE (A,25) Z2(1),C2(1),F2(1)
DO 200 I=2,N2
Z2(I)=Z2(I-1)+DX2
C2(I)=A24*SINH(P2*(Z2(I)-T2))+CA*COSH(P2*(Z2(I)-T2))
F2(I)=-AK2*P2*(A24*COSH(P2*(Z2(I)-T2))+CA*SINH(P2*(Z2(I)-T2)))
200 WRITE (A,25) Z2(I),C2(I),F2(I)
WRITE (A,27)
27 FORMAT ('0CONCENTRATION DISTRIBUTION IN SOIL.')
WRITE (A,31)
Z3(1)=0.0
C3(1)=A34*EXP(P3*Z3(1))
F3(1)=-AK3*P3*EXP(P3*Z3(1))*A34
WRITE (A,25) Z3(1),C3(1),F3(1)
DO 300 I=2,N3
Z3(I)=Z3(I-1)+DX3
C3(I)=A34*EXP(P3*Z3(I))
F3(I)=-AK3*P3*EXP(P3*Z3(I))
300 WRITE (A,25) Z3(I),C3(I),F3(I)
END

```

Appendix 6.1. Program BASFLUS, Distribution of Radon Concentrations and Fluxes Across Multilayered Porous Medium. Sealant Case.

```

PROGRAM BASFLUS(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)
DIMENSION Z1(10),Z2(10),Z3(10),C1(10),C2(10),C3(10),F1(10),F
2(10),F3(10)
SINH(X)=(EXP(X)+EXP(-X))/2.
COSH(X)=(EXP(X)-EXP(-X))/2.
READ(5,1)KA,RO,F
1 FORMAT(3E20.3)
WRITE(6,2)KA,RO,F
2 FORMAT(10HKA=*,E10.3,*GR KA/GK TAILIGS*,3X,*RO=*,E10.3,*GR/CM3*,3X,*
2F=*,E10.3)
READ(5,3)AK1,AK2,AK3
3 FORMAT(3E20.3)
WRITE(6,4)AK1,AK2,AK3
4 FORMAT(10HAK1=*,E10.3,*CM2/SEC*,3X,*AK2=*,E10.3,*CM2/SEC*,3X,*AK3=*,
2,E10.3,*CM2/SEC*)
READ(5,5)POR5,POR1,PORC
30 FORMAT(3E20.3)
WRITE(6,3)POR5,POR1,PORC
33 FORMAT(10HPOR5=*,E10.3,3X,*POR1=*,E10.3,3X,*PORC=*,E10.3)
READ(5,6)TH1,TH2
5 FORMAT(2E20.3)
READ(5,9)N1,N2,N3
9 FORMAT(3I10)
S=RA*RO*F*2.1E+06/POR1
P1=SQRT(2.1E+06/AK1*POR1)
P2=SQRT(2.1E+06/AK2*PORC)
P3=SQRT(2.1E+06/AK3*POR5)
D1=1./P1
D2=1./P2
D3=1./P3
TH3=7.*D3
WRITE(6,20)TH1,TH2,TH3
20 FORMAT(10HTH1=*,E10.3,3X,*TH2=*,E10.3,3X,*TH3=*,E10.3)
WRITE(6,11)S,D1,D2,D3
11 FORMAT(10HSOURCE=*,E10.3,*PC/CM3-SEC*,3X,*RELAXATION DISTANCES,*
2D1=*,E10.3,*CM*,3X,*D2=*,E10.3,*CM*,3X,*D3=*,E10.3,*CM*)
WRITE(6,10)N1,N2,N3
10 FORMAT(10H10*10*STEPS IN REGION 1,*,3X,10*STEPS IN REGION 2,*,3X,1
210*STEPS IN REGION 3,*)
TJ=TH1
T2=TH1+TH2
PT1=P1*T1
PT2=P2*T2
X1=SINH(PT1)
X2=COSH(PT1)
X3=S/2.1E+06
X4=AK3*P3
X5=-AK1*P1*COSH(PT1)
X6=AK1*P1*SINH(PT1)
X7=-COSH(P2*TH2)
X8=X3
X9=AK1*P1
X10=AK2*P2*SINH(P2*TH2)
O=X5*X10+X9*X4*X7-X4*(X1*X10+X9*X2*X7)
DA35=X3*(X5*X10+X9*X4*X7)+X8*X10*(X1*X6-X5*X2)
DA15=-X8*X6*X10-X4*X10*(X3-X4*X2)
DB15=X5*X4*X10-X4*(X1*X8*X10+X9*X3*X7)
DB25=X9*X6*X8-X4*X9*(X2*X8-X3)
A35=DA35/D
A15=DA15/D

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      B15=0.915/D
      B25=0.875/D
      WRITE(6,32) A15,B15,B25,A35
32  FORMAT('0A15=*,E10.3,*B15=*,E10.3,*B25=*,E10.3,*A35=*,E10.3)
      WRITE(6,21)
21  FORMAT('0FLUXES')
      F13=AK3*A35*P3
      F12=-AK1*A15*P1
      F5=0.0
      WRITE(6,22) F13,F12,F5
22  FORMAT('0F13=*,E10.3,3X,*F12=*,E10.3,3X,*F5=*,E10.3)
      WRITE(6,28)
28  FORMAT('0CONCENTRATION DISTRIBUTIONS.')
      DX1=TH1/(N1-1)
      DX2=TH2/(N2-1)
      DX3=TH3/(N3-1)
      WRITE(6,23) DX1,DX2,DX3
23  FORMAT('0STEP SIZES,DX1=*,F10.3,*CM*,3X,*DX2=*,F10.3,*CM*,3X,*DX3=
      2*,F10.3,*CM*')
31  FORMAT('0*,5X,*LOCATION*,17X,*CONCENTRATION*,17X,*FLUX*')
      WRITE(6,24)
24  FORMAT('0CONCENTRATION DISTRIBUTION IN THE TAILINGS.')
      WRITE(6,31)
      Z1(1)=0.0
      C1(1)=A15*S(N1*(P1*(Z1(1)-T1)))+B15*COSH(P1*(Z1(1)-T1))+5/2.1E-06
      F1(1)=-AK1*P1*(A15*COSH(P1*(Z1(1)-TH1))+B15*SINH(P1*(Z1(1)-TH1)))
      WRITE(6,25) Z1(1),C1(1),F1(1)
25  FORMAT('E15.5,10X,E15.5,10X,E15.5)
      DO 100 I=2,N1
      Z1(I)=Z1(I-1)+DX1
      C1(I)=A15*SINH(P1*(Z1(I)-T1))+B15*COSH(P1*(Z1(I)-T1))+5/2.1E-06
      F1(I)=-AK1*P1*(A15*COSH(P1*(Z1(I)-TH1))+B15*SINH(P1*(Z1(I)-TH1)))
100  WRITE(6,25) Z1(I),C1(I),F1(I)
      WRITE(6,26)
26  FORMAT('0CONCENTRATION DISTRIBUTION IN CONCRETE.')
      WRITE(6,31)
      Z2(1)=T1
      C2(1)=B25*COSH(P2*(Z2(1)-T2))
      F2(1)=-AK2*P2*B25*SINH(P2*(Z2(1)-T2))
      WRITE(6,25) Z2(1),C2(1),F2(1)
      DO 200 I=2,N2
      Z2(I)=Z2(I-1)+DX2
      C2(I)=B25*COSH(P2*(Z2(I)-T2))
      F2(I)=-AK2*P2*B25*SINH(P2*(Z2(I)-T2))
200  WRITE(6,25) Z2(I),C2(I),F2(I)
      WRITE(6,27)
27  FORMAT('0CONCENTRATION DISTRIBUTION IN SOIL.')
      WRITE(6,31)
      Z3(1)=0.0
      C3(1)=A35*EXP(P3*Z3(1))
      F3(1)=-AK3*P3*A35*EXP(P3*Z3(1))
      WRITE(6,25) Z3(1),C3(1),F3(1)
      DO 300 I=2,N3
      Z3(I)=Z3(I-1)+DX3
      C3(I)=A35*EXP(P3*Z3(I))
      F3(I)=-AK3*P3*A35*EXP(P3*Z3(I))
300  WRITE(6,25) Z3(I),C3(I),F3(I)
      END

```

Appendix 6.2. Program SLABCON. Gamma Exposure Dose from a Shielded Infinite Slab with Constant Volume Source Strength.

```

PROGRAM SLABCON(INPUT,OUTPUT,TAPEL=INPUT,TAPEH=OUTPUT)
DIMENSION FLU(30),DOSE(30),TMU(30),CMU(30),AMU(30),GAM(30),G(30),C
ZONV(30),SV(30),A(26,2),AL(26,2)
DATA(GAM(1:1)=1.26)/.609,.352,.295,.120,.1764,.1238,.769,.1378,.2
2204,.742,.14,.047,.935,.1728,.1509,.1848,.2445,.053,.2117,.51,.749
2,.803,.186,.26,.42,.61/
DATA(G(1:1)=1.26)/.47,.36,.19,.17,.17,.6,.5,.5,.5,.4,.4,.3,.3,.
22,.2,.2,.1,.1,.078,.014,.0011,.4,.007,.0002,.0002/
DATA(TMU(1:1)=1.26)/.174,.240,.288,.129,.102,.123,.150,.114,.078,.
2348,.114,.849,.138,.107,.108,.096,.03,.849,.084,.192,.15,.15,.299,
2.315,.217,.174/
DATA(CMU(1:1)=1.26)/.184,.23,.248,.138,.115,.129,.165,.122,.092,.2
271,.172,.984,.152,.115,.117,.106,.046,.984,.101,.198,.165,.165,.42
2,.264,.212,.184/
DATA(AMU(1:1)=1.26)/.091,.115,.125,.072,.058,.07,.084,.067,.048,.1
234,.067,.226,.077,.054,.062,.055,.024,.226,.053,.101,.084,.084,.15
2,.132,.11,.091/
DATA(A(1:1,1)=1.26)/11.75,13.30,13.75,9.25,7.0,8.8,10.95,8.3,5.8,
214,10.8,20.15,45.10,10.7,05.7,40.6,72.5,4.15,3.6,0,12.4,10.8,10.75
2,14.45,13.90,12.80,11.6/
DATA(A(1:2,1)=1.26)/-10.75,-12.30,-12.75,-8.25,-6.0,-7.8,-9.95,-7.
23,-4.4,-13.10,-7.29,-14.45,-9.10,-6.05,-6.40,-5.72,-4.4,-14.3,-5.0
2,-11.4,-9.4,-9.75,-13.45,-12.90,-11.80,-10.6/
DATA(AL(1:1,1)=1.26)/-.104,-.175,-.131,-.084,-.071,-.081,-.097,-.0
278,-.066,-.137,-.077,-.16,-.090,-.072,-.075,-.070,-.064,-.158,-.06
27,-.112,-.095,-.094,-.0143,-.134,-.118,-.103/
DATA(AL(1:2,1)=1.26)/0.014,0.002,0.030,.052,.037,.020,.042,.062,
2.0,.0425,0.027,.046,.045,.054,.066,0.060,.008,.021,.022,0.0,0,
2.005,.0145/
DATA(CONV(1:1)=1.26)/1.2,.7,.6,2.1,2.9,2.3,1.55,2.5,3.5,4.7,2.5,.1
23,1.8,2.9,2.65,1.1,3.6,.13,3.4,.13,1.55,.14,.33,.52,.85,1.2/
READ(5,1)RA,RO,E
1 FORMAT(3E20,3)
READ(5,2)PORT
2 FORMAT(E20,3)
READ(5,3)TT,TC,TA
3 FORMAT(3E20,3)
WRITE(6,5)RA,RO,E,PORT,TT,TC,TA
5 FORMAT('0RA=*E20,3/1RO*RO=*E20,3/1HO*E=*E20,3/1HO*PORT=*E20,3/
21HO*TT=*E20,3/1HO*TC=*E20,3/1HO*TA=*E20,3)
SC=RA*RO*1.E+17*3.7E-02
WRITE(6,8)SC
8 FORMAT('0SC=*E20,14)
DO 10 I=1,26
FLU(I)=0.0
SV(I)=SC*G(I) *1.E-02
DO 99 J=1,2
DO 99 K=1,2
BTMU=TMU(I)*(1.+AL(I,J))
BCMU =CMU(I)*(1.+AL(I,K))
B1=BCMU *TC+AMU(I)*TA*1.E-03
B3=B1+BTMU *TT
WRITE(6,40)B1,B3
40 FORMAT('00*E10,3,5X*E10,3)
E1=E2(B1)
E3=E2(B3)
99 FLU(I)=FLU(I)+A(I,J)*A(I,K)*(E1)-E3)/BTMU
FLU(I)=FLU(I)*SV(I)/2,

```

```

DOSE(I)=FLU(I)*CONV(I)*1.E-06
10 WRITE(6,6)GAM(I),G(I),TMU(I),CMU(I),AMU(I),FLU(I),DOSE(I)
6 FORMAT(' 4.7E19.10)
FLUTOT=FLU(I)
WRITE(6,13)
13 FORMAT('CONTRIBUTION FROM RADON IN TAILINGS PARTICLES. ')
DO 11 I=2,22
11 FLUTOT=FLUTOT+FLU(I)
DOSETOT=DOSE(I)
DO 12 I=2,22
12 DOSETOT=DOSETOT+DOSE(I)
FLUP1=FLUTOT*(1.-E)
DOSEP1=DOSETOT*(1.-E)
WRITE(6,14)FLUP1,DOSEP1
14 FORMAT('0FLUP1=*E20.5,*PHOTONS/CM2.SEC*/1H0,*DOSEP1=*E20.5,*R/HR
2. *)
WRITE(6,15)
15 FORMAT('0CONSTANT COMPONENT OF RADON PROFILE. ')
FLUP2=FLUTOT*F
DOSEP2=DOSETOT*E
WRITE(6,16)FLUP2,DOSEP2
16 FORMAT('0FLUP2=*E20.5,*PHOTONS/CM2.SEC*/1H0,*DOSEP2=*E20.5,*R/HR
2. *)
WRITE(6,17)
17 FORMAT('0CONTRIBUTION FROM RADIUM IN TAILINGS. ')
FLUP3=0.0
DOSEP3=0.0
DO 18 I=23,26
18 FLUP3=FLUP3+FLU(I)
DO 19 I=23,26
19 DOSEP3=DOSEP3+DOSE(I)
WRITE(6,20)FLUP3,DOSEP3
20 FORMAT('0FLUP3=*E20.5,*PHOTONS/CM2.SEC*/1H0,*DOSEP3=*E20.5,*R/HR
2. *)
END
FUNCTION E2(X)
IF(1.-X)101,101,102
101 E2 =EXP(-X)*(1.-10.2372905+4.5307924*X+5.1266902*X*X+X*X*X)/(
22.4766731+8.6660126*X+6.1255272*X*X+X*X*X)
GO TO 103
102 E2 =EXP(-X)-X*(-0.57721566-ALOG(X)+X-X**2/4.+X**3/18.-X**4/96.+X
2**5/600.-X**6/4320.+X**7/35280.-X**8/322560.)
103 CONTINUE
RETURN
END

```


Appendix 6.3. Program SLAPROT. Gamma Exposure Dose from a Shielded Infinite Slab Source of Tailings with Variable Source Strength. Sealant and No-Sealant Cases.

```

PROGRAM SLAPROT(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)
DIMENSION FLU(30),DOSE(30),THU(30),CHU(30),AMU(30),GAM(30),b(30),C
ZONV(30),A(26,2),AL(26,2),FLU1(30),FLU2(30),SV1(30),SV2(30),
2X(10)
DATA(GAM(1),I=1,26)/.609,.752,.295,1.120,1.764,1.238,.769,.378,.2,
2204,.242,1.4,.047,.435,1.728,1.509,1.848,2.445,.053,2.117,.51,.799
2,.803,.186,.26,.42,.61/
DATA(G(1),I=1,26)/47,.36,.19,.17,.17,.6,.5,.5,.5,.4,.4,.3,.3,
22,.2,.2,.1,.1,.078,.014,.0011,.4,.007,.0002,.0002/
DATA(THU(1),I=1,26)/.174,.246,.288,.129,.102,.123,.150,.114,.078,.
2348,.114,.49,.138,.102,.108,.096,.03,.849,.084,.192,.15,.15,.299,
2,315,.217,.174/
DATA(CHU(1),I=1,26)/.194,.23,.248,.138,.115,.129,.165,.122,.092,.2
271,.127,.98,.152,.115,.117,.106,.046,.984,.101,.198,.165,.165,.42
2,.264,.217,.194/
DATA(AMU(1),I=1,26)/.091,.115,.125,.072,.058,.07,.084,.067,.048,.1
234,.067,.226,.077,.058,.062,.055,.074,.226,.053,.101,.084,.084,.15
2,.132,.11,.091/
DATA(A(1,1),I=1,26)/11.75,13.30,13.75,9.25,7.0,8.8,10.95,8.3,5.8,
214,10.8,20,15.45,10.10,7.05,7.60,6.72,5.4,15.3,6.0,12.4,10.8,18.75
2,14.45,13.90,12.80,11.6/
DATA(A(1,2),I=1,26)/-10.75,-12.30,-12.75,-8.25,-6.0,-7.8,-9.95,-7,
23,-4.8,-13.10,-7.70,-14.45,-9.10,-6.05,-6.80,-5.72,-4.4,-14.3,-5.0
2,-11.4,-9.8,-9.75,-13.45,-12.90,-11.80,-10.6/
DATA(AL(1,1),I=1,26)/-.104,-.125,-.131,-.084,-.071,-.081,-.097,-.0
278,-.066,-.137,-.077,-.14,-.090,-.072,-.075,-.070,-.064,-.158,-.06
27,-.112,-.095,-.044,-.0143,-.134,-.118,-.103/
DATA(AL(1,2),I=1,26)/0.014,0.002,0.0,0.030,0.052,0.037,0.020,0.042,0.062,
2,0.0425,0.0,0.027,0.048,0.045,0.054,0.066,0.0,0.060,0.008,0.021,0.022,0.0,0,
2,005,0.0145/
DATA(CONV(1),I=1,26)/1.2,.7,0.6,2,1.2,9,2.3,1.55,2.5,3.5,4.7,2.5,.1
23,1.8,2.9,2.65,3,1,3.0,13,3.4,13,1.55,14,33,52,85,1,2/
READ(5,1)RA,R0,E
1 FORMAT(3E20,3)
READ(5,2)PORT,PORC
2 FORMAT(2E20,3)
READ(5,4)D1,D2,D3
4 FORMAT(3E20,3)
READ(5,3)TT,TC,TA
3 FORMAT(3E20,3)
READ(5,5)A14,B14,A15,B15
5 FORMAT(4E20,3)
WRITE(6,8)RA,R0,E,PORT,PORC,TT,TC,TA
8 FORMAT('0RA=',E20,3/1H0'R0=',E20,3/1H0'E=',E20,3/1H0'PORT=',E20,3/
21H0'PORC=',E20,3/1H0'TT=',E20,3/1H0'TC=',E20,3/1H0'TA=',E20,3)
WRITE(6,9)D1,D2,D3
9 FORMAT('0D1=',E20,4,3X,'D2=',E20,4,3X,'D3=',E20,4)
WRITE(6,36)A14,B14,A15,B15
36 FORMAT('0A14=',E20,3,3X,'B14=',E20,3,3X,'A15=',E20,3,3X,'B15=',E20
2,3)
XK1=1./D1
XK2=XK1
DO 100 L=1,2
IF(L.EQ.2)GO TO 101
S11=(A14+B14)*EXP(-XK1*TT)/2.*PORT
S12=(B14-A14)*EXP(XK1*TT)/2.*PORT
GO TO 102
101 S11=(A15+B15)*EXP(-XK1*TT)/2.*PORT
S12=(B15-A15)*EXP(XK1*TT)/2.*PORT

```

```

102 WRITE(6,7)S11,XK1,S12,XK2
71 FORMAT(*0S11=*E20.10,3X,*XK1=*E20.10,3X,*S12=*E20.10,3X,*XK2=*E20.10)
    IF(4.E0.2)GO TO 103
    WRITE(6,31)
31 FORMAT(*0CONCENTRATION PROFILE IN THE TAILINGS,NO SEALANT.*)
    GO TO 104
103 WRITE(6,34)
34 FORMAT(*0CONCENTRATION PROFILE IN THE TAILINGS,WITH SEALANT.*)
104 SP=RA*ROE*1.E-12/PORT
    DX=TT/6.
    WRITE(6,35)
35 FORMAT(*0*,6X,*LOCATION*,12X,*TERM1*,16X,*TERM2*,16X,*CONCENTRATION*,2NS.*)
    DO 30 M=1,7
    X(M)=(M-1)*DX
    TERM1=S11/PORT*EXP(XK1*X(M))
    TERM2=S12/PORT*EXP(XK2*X(M))
    PROF=TERM1+TERM2*SP
30 WRITE(6,32)X(M),TERM1,TERM2,PROF
32 FORMAT(*0*,9X,E21.5,3X,E21.5,3X,E21.5,3X,E21.5)
    WRITE(6,33)SP
33 FORMAT(*0SP=*E21.14)
    DO 10 I=1,22
    SV1(I)=S11*(1+1.E-02*3.7E-02
    SV2(I)=S12*(1+1.E-02*3.7E-02
    FLU1(I)=0.0
    FLU2(I)=0.0
    DO 99 J=1,2
    DO 99 K=1,2
    BTMU=TMU(I)*(1+AL(I,J))
    BCMU=CMU(I)*(1+AL(I,K))
    B1=BCMU*TC*AMU(I)*TA*,E-03
    H3=B1+BTMU*TT
    XKOMU1=-XK1/BTMU
    XKOMU2=-XK2/BTMU
    ARG1=B1*(1.-XKOMU1)
    ARG2=B3*(1.-XKOMU1)
    EB1=EXP(B1*XKOMU1)*F(H1)+F(ARG1)
    EB3=EXP(B3*XKOMU1)*F(H3)+F(ARG2)
    FLUA=A(I,K)*EXP(XK1*H3/BCMU)*(EB3-EB1)/BCMU/XKOMU1*A(I,J)
    FLU1(I)=FLU1(I)+FLUA
    ARG3=B1*(1.-XKOMU2)
    ARG4=B3*(1.-XKOMU2)
    EB1=EXP(B1*XKOMU2)*F(H1)+F(ARG3)
    EB3=EXP(B3*XKOMU2)*F(H3)+F(ARG4)
    FLUA=A(I,K)*EXP(XK2*H3/BCMU)*(EB3-EB1)/BCMU/XKOMU2*A(I,J)
99 FLU2(I)=FLU2(I)+FLUA
    FLU(I)=FLU1(I)*SV1(I)/2.
    FLU2(I)=FLU2(I)*SV2(I)/2.
    FLU(I)=FLU1(I)+FLU2(I)
    DOSE(I)=FLU(I)*CONV(I)*1.E-06
10 WRITE(6,6)GAX(I),G(I),TMU(I),CMU(I),AMU(I),FLU(I),DOSE(I)
6 FORMAT(*,7E19,10)
    FLUTOT=FLU(I)
    DO 11 I=2,22
11 FLUTOT=FLUTOT+FLU(I)
    DOSETOT=DOSE(I)
    DO 12 I=2,22
12 DOSETOT=DOSETOT+DOSE(I)
    WRITE(6,7)FLUTOT,DOSETOT
7 FORMAT(*0FLUTOT=*E20.14,*PHOTONS/CM2.SEC/*,DOSETOT=*E20.14,*R/H
2R.*)
100 CONTINUE
    END

```

```

      FUNCTION F(Y)
      IF(Y)202,201,201
201 IF(1.-Y)101,101,102
101 F = EXP(-Y)* 10.2372905+.5307924*Y+.5.1266902*Y*Y+Y**3 )/(
      22.4766331+.8.6660126*Y+.6.1255272*Y*Y+Y**3 )/Y
      GO TO 103
102 F =-0.57721566+ALOG(Y)+Y-Y**2/4.+Y**3/18.-Y**4/96.+Y**5/600.-Y**
      + 26/4320.+Y**7/35280.-Y**8/322560.
      GO TO 103
202 Y=-Y
      F=-10.57721566+ALOG(Y)+Y+Y**2/4.+Y**3/18.+Y**4/96.+Y**5/600.+Y**6/
      24320.+Y**7/35280.+Y**8/322560.)
103 CONTINUE
      RETURN
      RETURN
      END

```

Appendix 6.4. Program SLAPROC. Gamma Exposure Dose from a Shielded Infinite Slab of Concrete with Variable Source Strength. Sealant and No-Sealant Cases.

```

PROGRAM SLAPROC(INPUT,OUTPUT,TAPL=(INPUT,TAPF=OUTPUT)
DIMENSION FLU(30),DOSE(10),CHU(30),TMU(30),AMU(30),GAM(30),G(30),C
20NV(30), A(26*2),AL(26*2),FLU1(30),FLU2(30),SV1(30),SV2(30),
2X(10)
DATA(GAM(I),I=1,26)/.609,.352,.295,.1.120,.1.764,.1.238,.769,.1.378,.2.
2204,.242,.1.4,.047,.935,.1.728,.1.504,.1.848,.2.445,.053,.2.117,.51,.799
2,.803,.185,.26,.42,.61/
DATA(G(I),I=1,26)/.47,.36,.19,.17,.17,.6,.5,.5,.4,.4,.3,.3,.
22,.2,.2,.1,.1,.078,.014,.001,.4,.007,.0002,.0002/
DATA(TMU(I),I=1,26)/.174,.740,.280,.124,.102,.123,.150,.114,.078,.
2348,.114,.449,.138,.102,.108,.096,.03,.849,.084,.192,.15,.15,.299,
2.315,.717,.174/
DATA(CHU(I),I=1,26)/.144,.23,.248,.138,.115,.129,.165,.122,.092,.2
271,.122,.984,.152,.115,.117,.106,.046,.984,.101,.198,.165,.165,.42
2,.264,.212,.194/
DATA(AMU(I),I=1,26)/.09,.115,.125,.072,.058,.07,.084,.067,.048,.1
234,.067,.226,.077,.058,.067,.055,.024,.226,.053,.101,.084,.084,.15
2,.132,.11,.091/
DATA(A(I,1),I=1,26)/11.75,13.30,13.75,9.25,7.0,8.8,10.95,8.3,5.8,
214.10,4.20,15.45,10.10,7.05,7.90,6.72,5.4,15.3,6.0,12.4,10.8,10.75
2,14.45,13.90,12.80,11.6/
DATA(A(I,2),I=1,26)/-10.75,-12.30,-12.75,-8.25,-6.0,-7.8,-9.95,-7.
23,-4.8,-13.10,-7.20,-14.45,-4.10,-6.05,-6.80,-5.72,-4.4,-14.3,-5.0
2,-11.4,-9.8,-9.75,-13.45,-12.90,-11.80,-10.6/
DATA(AL(I,1),I=1,26)/-.134,-.175,-.131,-.084,-.071,-.081,-.097,-.0
278,-.066,-.137,-.077,-.16,-.040,-.072,-.075,-.070,-.064,-.158,-.06
27,-.117,-.095,-.094,-.0143,-.134,-.115,-.103/
DATA(AL(I,2),I=1,26)/0.014,0.002,.0,.030,.052,.037,.020,.042,.062,
2,0,.0425,.0,.027,.046,.045,.054,.066,.0,.060,.008,.021,.022,.0,.0,
2.005,.0145/
DATA(CONV(I),I=1,26)/1.2,2.7,6.2,1.2,2.9,2.3,1.55,2.5,3.5,4.7,2.5,1
23,1.8,2.9,2.65,3.1,3.6,.13,3.4,.13,1.55,.14,.33,.52,.85,1.2/
READ(5,1)KA,K,E
1 FORMAT(3E20.3)
READ(5,2)PORT,PORC
2 FORMAT(2E20.3)
READ(5,4)D1,D2,D3
4 FORMAT(3E20.3)
READ(5,7)TT,TC,TA
3 FORMAT(3E20.3)
READ(5,5)A24,CA,H25
5 FORMAT(3E20.3)
WRITE(16,8)KA,QA,E,PORT,PORC,TT,TC,TA
8 FORMAT('044=*,E20.3/1H0*K0=*,E20.3/1H0*E=*,E20.3/1H0*PORT=*,E20.3/
21H0*PORC=*,E20.3/1H0*TT=*,E20.3/1H0*TC=*,E20.3/1H0*TA=*,E20.3)
WRITE(6,70)D1,D2,D3
70 FORMAT('001=*,E20.4,3X,*D2=*,E20.4,3X,*D3=*,E20.4)
WRITE(6,36)A24,CA,H25
36 FORMAT('0424=*,E10.3,3X,*CA=*,E10.3,3X,*H25=*,E10.3)
KK1=1./D2
KK2=KK1
DO 100 L=1,2
IF(L.EQ.2)GO TO 101
S13=(A24*CA)*EXP(-KK1*TC)/2.
S14=(CA-A24)*EXP(KK1*TC)/2.
GO TO 102
101 S13=H25*EXP(-KK1*TC)/2.
S14=H25*EXP(KK1*TC)/2.
102 WRITE(16,71)S13,KK1,S14,KK2
71 FORMAT('0513=*,E20.10,3X,*KK1=*,E20.10,3X,*S14=*,E20.10,3X,*KK2=*,
2E20.10)

```

```

      IF IL.EQ.2160 TO 103
      WRITE(6,31)
31  FORMAT('CONCENTRATION PROFILE IN THE CONCRETE.WO SEALANT.')
```

GO TO 104

```
103 WRITE(6,34)
34  FORMAT('CONCENTRATION PROFILE IN THE CONCRETE.WITH SEALANT.')
```

104 DX2=TC/6.

```
      WRITE(6,35)
35  FORMAT('0*.6X.*LOCATION*.17X.*TERM1*.16X.*TERM2*.16X.*CONCENTRATIO
2NS.')
```

DO 30 M=1,7

```
      X(M)=(M-1)*DX2
      TERM1=S13 *EXP(XK1*X(M))
      TERM2=S14 *EXP(XK2*X(M))
      PROF=TERM1+TERM2
```

30 WRITE(6,32)X(M),TERM1,TERM2,PROF

```
32  FORMAT('0*.4X.E21.5 .3X.E21.5 .3X.E21.5 .3X.E21.5 ')
      DO 10 I=1,22
      SV1(I)=S13*G(I)*1.E-02*3.7E-02*PWRG
      SV2(I)=S14*G(I)*1.E-02*3.7E-02*PWRG
      FLU1(I)=0.0
      FLU2(I)=0.0
      DO 99 K=1,2
      BCMU =CMU(I)*(1.+AL(I,K))
      B1=AMU(I)*TA*1.E-03
      B3=B1+PCMU*TC
      XKOMU1=-XK1/BCMU
      XKOMU2=-XK2/BCMU
      ARG1=B1*(1.-XKOMU1)
      ARG2=B3*(1.-XKOMU1)
      EB1=EXP(B1*XKOMU1)*F(H1)*F(ARG1)
      EB3=EXP(B3*XKOMU1)*F(H3)*F(ARG2)
      FLUA=A(I,K)*EXP(XK1*H1/BCMU)*(EB3-EB1)/BCMU/XKOMU1
      FLU1(I)=FLU1(I)+FLUA
      ARG3=B1*(1.-XKOMU2)
      ARG4=B3*(1.-XKOMU2)
      EB1=EXP(B1*XKOMU2)*F(H1)*F(ARG3)
      EB3=EXP(B3*XKOMU2)*F(H3)*F(ARG4)
      FLUA=A(I,K)*EXP(XK2*H3/BCMU)*(EB3-EB1)/BCMU/XKOMU2
99  FLU2(I)=FLU2(I)+FLUA
      FLU1(I)=FLU1(I)*SV1(I)/2.
      FLU2(I)=FLU2(I)*SV2(I)/2.
      FLU(I)=FLU1(I)+FLU2(I)
      DOSE(I)=FLU(I)*CONV(I)*1.E-06
10  WRITE(6,6)GAM(I),G(I),THU(I),CMU(I),AMU(I),FLU(I),DOSE(I)
6  FORMAT('*.*,?E19.10)
      FLUTOT=FLU(I)
      DO 11 I=2,22
11  FLUTOT=FLUTOT+FLU(I)
      DOSETOT=DOSE(I)
      DO 12 I=2,22
12  DOSETOT=DOSETOT+DOSE(I)
      WRITE(6,7)FLUTOT,DOSETOT
7  FORMAT('0FLUTOT=*.E20.14,*PHOTONS/CM2.SEC*/ *DOSETOT=*.E20.14,*R/H
2R.')
```

100 CONTINUE

END

```

      FUNCTION F(Y)
      IF(Y)202,201,201
201  IF(1.-Y)101,101,102
101  F = EXP(-Y)* (0.2372405+4.5307924*Y+5.1266902*Y*Y+Y**3 )/1
      22.4764331+8.6660126*Y+6.1245272*Y*Y+Y**3 )/Y
      GO TO 103
102  F =-0.57721566-ALOG(Y)+Y-Y**2/4.+Y**3/18.-Y**4/96.+Y**5/600.-Y**
      26/4320.+Y**7/35280.-Y**8/322560.
      GO TO 103
202  Y=-Y
      F=- (0.57721566+ALOG(Y)+Y-Y**2/4.+Y**3/18.-Y**4/96.+Y**5/600.+Y**6/
      24320.+Y**7/35280.+Y**8/322560.)
103  CONTINUE
      RETURN
      END

```